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Power Plant Stack Plumes in Complex Terrain: An Appraisal of Current Research

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POWER PLANT STACK PLUMES IN COMPLEX TERRAIN: An Appraisal of Current Research

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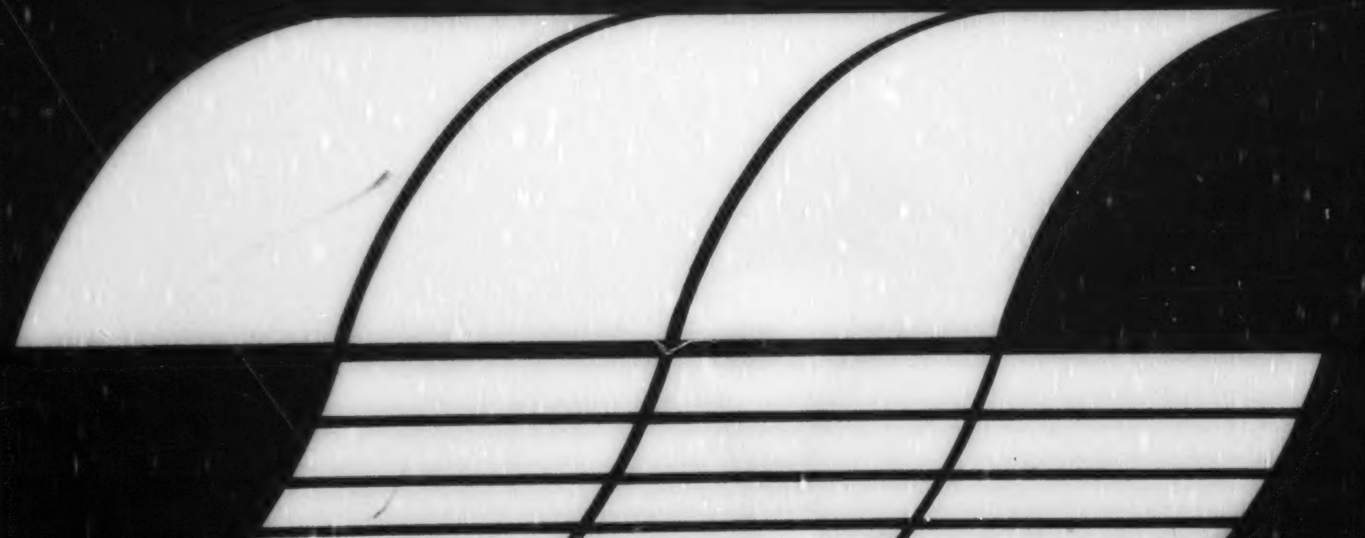

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POWER PLANT STACK PLUMES IN COMPLEX TERRAIN

An Appraisal of Current Research

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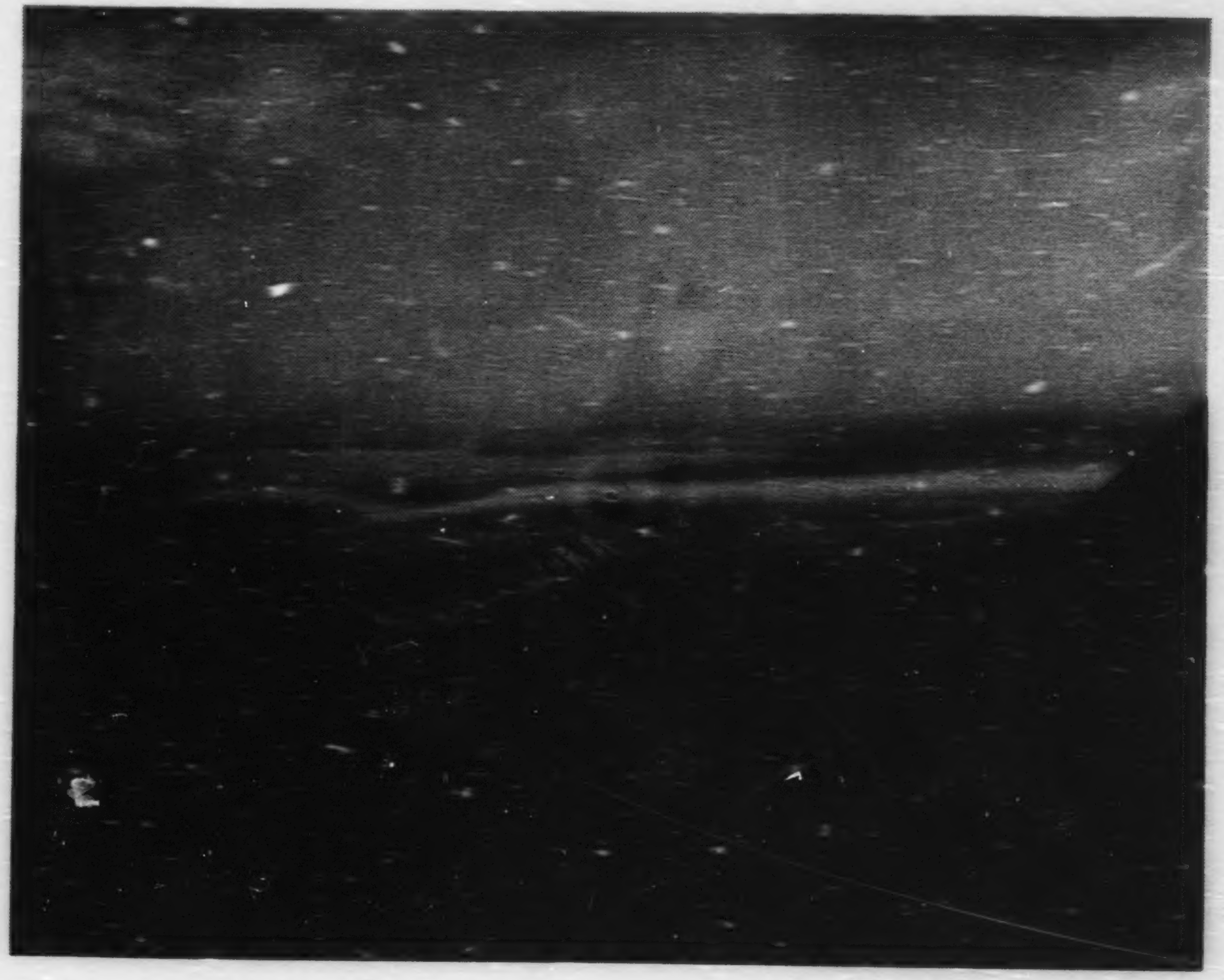
ABSTRACT

This report reviews the literature of scientific studies of the behavior of stack plumes from fossil-fueled electric power plants in complex (hilly or mountainous) terrain. Non-conservative chemical transformation and depletion, and conservative transport and diffusion of pollutants, are considered. Studies of SO_2 oxidation rates in power plant plumes are described and the primary mechanisms for conversion to sulfate are detailed. Homogeneous gas phase reaction of SO_2 with OH is of importance, with conversion rates reported from 1 to 13 percent per hour. Aqueous phase SO_2 oxidation has higher conversion rates, in presence of NH_3 and with iron, lead or manganese as catalysts, but is less well-studied. Early depletion of background ozone by stack plumes is documented; evidence indicates that ozone above background levels reappears in plumes at great distances from the stack. Little information on field studies of plume chemistry of NO_x, CO, or hydrocarbons was found. Scavenging of SO_2 from plumes by precipitation is shown to increase with distance from the source and has been modeled. Surface contact may be an important plume depletion process but quantitative models have not been validated.

Current theories of airflow, turbulence and diffusion phenomena in complex terrain are described, and are exemplified through the review of sixteen field observation programs of the physical behavior of plumes from continuous elevated sources in complex terrain. The review discusses program objectives, data sampling methodologies, model-to-measurement comparisons associated with the field programs and program conclusions. The types of models available to simulate plume behavior numerically are discussed in the contexts of these programs. In addition, results are reported of an independent GEOMET analysis of σ_y and σ_z relationships in the Gaussian plume model over complex terrain, using data from LAPPES and TVA field measurement programs.

Results of the review indicate that turbulence is enhanced over complex terrain through thermal drainage, up-slope flows, lee waves and channeling. Evidence exists that horizontal spreading of plumes is amplified in complex terrain to a greater extent than vertical spreading. Available models tend to overpredict plume concentrations by factors of 5 or more at distances within a few kilometers of the source but prediction performance improves at greater distances. The models in general underestimate the initial lateral spread of the plume and overestimate vertical spread at greater distances.



A view of the plume from the Four Corners Power Plant in New Mexico, crossing the Hogback and going through two lee-wave oscillations. The plant is situated to the east (right) of this photograph.

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PART 1

SCOPE OF EFFORT AND RESULTS

Section I

INTRODUCTION

At the end of October, 1975, the Meteorology and Assessment Division of the U.S. Environmental Protection Agency, at Research Triangle Park, North Carolina, awarded GEOMET, Incorporated a contract to undertake a study - including literature research, field observations, and analytic modeling - into the behavior of stack plumes from large fossil-fueled power plants in complex terrain. The first product of that study, an appraisal of the extent, utility and limitations of existing knowledge in this field, is the subject of this report.

This report is the culmination of Phase I of GEOMET's work under EPA Contract Number 68-02-2260. A Phase II report, describing results of a long-term program for field monitoring of sulfur oxides and other plume components begun by GEOMET in the vicinity of the 700 MW coal-fired Clinch River Power Plant at Carbo, Virginia in April, 1976, will be issued in late 1977.

BACKGROUND

Fossil-fueled electric power plants are major sources of air pollution. Particulate matter, sulfur dioxide, nitrogen oxides, and trace metals are - in the absence of effective control equipment - dispersed into the air from power plant stacks. All can be hazardous to health and welfare in the plant environs. Chemical reactions of pollutants in power plant plumes may result in formation of sulfates, nitrates, and photochemical oxidants which are advected over great distances, resulting in health hazards in areas remote from the power plant source of the pollution. Permissible maximum ambient concentrations of sulfur dioxide (SO_2), nitrogen oxides (NO_x), total suspended particulate (TSP), and oxidants (O_x) have been established in law and are enforced under State Air Quality Control Implementation Plans. National standards for ambient sulfate and nitrate concentrations do not yet exist, but may be expected in the future.

Particulate matter emissions can be effectively controlled at large power plants through electrostatic precipitation. Sulfur dioxide emission control procedures are technically feasible but are expensive; they have not been widely adopted. The electric power industry has typically relied upon the dispersive power of the atmosphere to reduce ambient ground level SO_2 concentrations, through discharge of emissions from tall stacks. Effective means for the control of NO_x emissions are still in development, not yet applicable to large-scale commercial production of electricity.

Growing reliance upon coal as a fuel for power generation - switching from imported oil and scarce gas - has increased the national concern about the SO_2 and sulfate air pollution problem. Much of the coal available in the United States has a relatively high sulfur content (more than one percent), but this fuel is our most sure source of future energy for many years to come.

Coal and oil-fired power plants were originally built near load centers, typically in or near urban areas. With the development of improved high voltage, long distance power transmission systems it has become economical to build very large (1000 MW and more) coal-fired power plants in coal mining areas, transmitting power hundreds of miles to load centers. Power plants in such remote areas are often located in complex mountainous terrain, in which ground elevations as high or higher than the top of plant stacks occur within the area traversed by the pollutant plumes from the plant. This condition may also occur in hilly, settled country, particularly where smaller or older power plants have stacks less than about 150 m in height.

For the design and siting of new power plants, and for the control of pollutants from existing ones, it is necessary to have reliable techniques for calculating the ambient air quality impact of stack emissions from large power plants - particularly for SO_2, NO_x and their transformation products. The problem has been studied in depth for many years. A body of modeling technology exists, suitable for calculating the physical behavior of a stack plume based upon input knowledge of pollutant emissions, stack configuration, and meteorology - but the models perform well only under limited conditions of terrain uniformity; and they do not perform well for non-conservative, reactive pollutants.

Existing models generally assume a uniform terrain roughness. Some models take differential terrain elevations into account, with broad simplifications and limited success. Models available to planners are typically weak (or totally lacking) in ability to handle plume depletion due to chemical transformation and non-conservative behavior.

A need exists to develop improved techniques for calculating the behavior of pollutants in the plumes of large power plants in complex terrain. Realistic goals for new studies can only be established through an examination of the extent, capability, and limitations of the existing body of knowledge. That examination is the subject of this report.

OBJECTIVES

The study which produced this report had the following objectives:

- Provision of an up-to-date description of existing knowledge of (1) concentrations of SO_2 and sulfate (and pollutants interacting with these), resulting from the operations of large fossil-fueled power plants in complex terrain, (2) relationships between meteorological data, terrain characteristics, and plume behavior, and (3) the pollutant transformation and depletion.
- Appraisal of the adequacy of typical meteorology, air quality and emissions data - of kinds routinely available to power plant plume analysts in non-research

situations - for use in calculating ground level concentrations of SO_2 and sulfates, due to power plant emissions, in areas of complex terrain.

Implicit in these objectives are the concepts of identifying deficiencies in current knowledge and recommending new research which could improve our ability to calculate the air pollution impact of power plant plumes in complex terrain.

Precise definitions of "large power plant" or of "complex terrain" are difficult to outline. In general, a large power plant may be thought of as one in which more than 700 MW of capacity are installed at one location. For plume study purposes, the complexity of terrain is a function of its influence upon plume behavior. As a working definition, complex terrain is terrain in which stack pollutants appear at ground surfaces in concentrations significantly different from those which would be expected to occur if the terrain were essentially level.

APPROACH

The study was essentially performed in a six-month period, from November, 1975 through April, 1976. The method was primarily that of a literature search, supplemented by discussions with researchers active in related areas. Research literature was sought out, reviewed, summarized, and appraised in the two major categories of (1) chemical transformations in power plant plumes and (2) the concentrations of pollutants in plumes moving under meteorological influences over complex terrain. In these categories information was sought on field observations of relevant data and also upon analytic efforts to develop, improve, and validate models for calculating pollutant behavior. GEOMET made some independent modeling analyses of SO_2 and meteorological data taken by others in previous field studies. In these modeling analyses GEOMET sought preliminary insights into relationships between terrain complexity and variations in the horizontal and vertical standard deviations (σ_y and

σ_z) of pollutant distribution models conventionally used to describe the dispersion of a plume. The results have been used to suggest further directions for study.

Section II

SUMMARY AND CONCLUSIONS

Over 200 studies, directly or indirectly relating to the subject of plume behavior from elevated point sources in complex terrain, were reviewed for the preparation of this report. The titles and authors of these studies are listed in an appended Bibliography. Aspects of plume behavior have been broadly categorized into two areas of information, namely, (1) non-conservative aspects relating to chemical transformation and depletion processes and (2) transport and diffusion aspects.[*]

Information presented regarding plume chemistry includes a discussion of the principles and results of investigative laboratory work for each of four classes of sulfate formation processes, a review of the approaches which have been used in plume chemistry modeling, and descriptions of major field observation programs. Other information presented regarding plume depletion processes includes a discussion of the principles of precipitation scavenging and chemical processes other than sulfate formation, modeling approaches for treating wet and dry deposition, and a description of major field observation programs involving precipitation scavenging measurements. An evaluation of the current state-of-the-art knowledge of the non-conservative aspects of plume behavior follows the discussions of principles, modeling, and field programs.

With regard to transport and diffusion aspects of plume behavior, the principles and observations of airflow and turbulence and diffusion enhancement in complex terrain are reviewed, followed by a discussion of the modeling of terrain influence on plume behavior and by detailed descriptions of 16 major field studies. An evaluation is made of the current state-of-the-art knowledge of the transport and diffusion of plumes under the influence of complex terrain.

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[*] In this review we have used transport, diffusion and dispersion with the following meanings: Transport - Movement of plume components by winds, at scales large enough to be treated by the equation of motion; Diffusion - Exchange of plume components and other atmospheric elements between regions in space, in apparently random motions, at a scale too small to be treated by the equations of motion; Dispersion - The combined result of the transport and diffusion processes.

Based on the review of previous studies of plume behavior and on a new analysis of the behavior of plumes from three power plants located in the Appalachian mountain regions of Pennsylvania and in Tennessee, we have drawn conclusions regarding the present state-of-the-art and needs for further research to treat plume behavior from a coal-fired power plant in complex terrain. Conclusions reached regarding the current state-of-the-art of non-conservative plume processes are presented first. The findings concern available measurements, modeling, sulfate conversion mechanisms, and plume depletion-deposition processes. The conclusions reached with regard to plume transport and diffusion then follow.

PLUME CHEMISTRY

A wide range of SO_2 oxidation rates (varying from zero to over 50 percent per hour) have been deduced from field study measurements. The rates reflect variations in meteorology (especially relative humidity), time of year, location, source emissions, and background atmospheric contaminants; most of these variables cannot be controlled in field experiments. A field program design that can serve as a guide in planning future studies is needed. Normally plume measurements show a depletion of ozone below the normal background concentration. However, levels greater than background have been evident in several studies of plant plumes at distances in excess of 50 km from the plume source. Current and future field programs are being designed to determine if ozone is formed in plumes under certain conditions. Very limited field observations have been made of chemical processes (other than the SO_2 to sulfate conversion) which involve ozone, NO, NO_2, CO, hydrocarbons, water, and radical chemistry.

Although several plume chemistry models have been developed, there is a definite lack of data to test these models. Additional research is needed in the areas of plume physics, plume photochemistry, and plume measurement methodology. Improved information on the dominant SO_2 conversion mechanisms and their reaction rates are required for realistic depiction of plume chemical processes.

Different SO_2 oxidation mechanisms may be important in power plant plumes under different environmental conditions. The significance of different conditions can only be hypothesized at this time. The four major processes of sulfate formation that have been identified from theoretical considerations to be potentially active in plant plumes include homogeneous indirect photo-oxidation, homogeneous air oxidation in liquid droplets, quasi-homogeneous catalyzed oxidation in liquid droplets and heteorogeneous catalyzed oxidation on dry surfaces.

The gas phase reaction of SO_2 with OH is the most notable homogeneous sulfate formation process. An SO_2 sulfate conversion rate of 2 percent hr^{-1} is a representative average rate for this reaction. Further quantification of the importance of the homogeneous OH-SO_2 mechanism compared to heterogeneous processes can be determined with more complete information on OH atmospheric profiles. SO_2 conversion rates for homogeneous gas phase reactions vary from 1 to 13 percent hr^{-1}.

Sulfate conversion rates of 0.1 to 2.0 percent per minute have been reported in studies of aqueous phase SO_2 oxidation. The process rate is highly variable due to its sensitivity to several parameters including temperature, relative humidity, and pH. The important role of NH_3 in promoting this conversion by maintaining a high pH has been documented. Available information for measuring NH_3 in the short time frames and low concentrations typical of airborne plume measurements is not adequate for field studies; similar problems exist with regard to field monitoring of the roles of trace metal catalysts.

Various transition metal ions (e.g., Mn^{++}, Cu^{++}, Fe^{++}, and Co^{++}) have a catalytic effect on the conversion of SO_2 to sulfate by a wide variety of heterogeneous reactions in solution conditions. Studies point to SO_2 conversion rates from 0 to 6 percent hr^{-1}, with the most efficient catalysts being lead and iron. Research has shown dry particulate vanadium to be relatively inactive as an effective catalyst in the heterogeneous oxidation of SO_2.

PLUME DEPLETION

The few field studies of precipitation scavenging of reactive pollutants from power plant plumes have measured the washout process by examining ground rainwater samples. The most comprehensive and recent study at the Centralia power plant in Washington showed that scavenging rates of SO_2 increase with distance from the plant, and that sulfate concentrations in rainwater due to the plume presence are not significant less than 10 km from the stack. The Scavenging Model Incorporating Chemical Kinetics (SMICK) has been developed and has the capability to predict precipitation scavenging of reactive pollutants from power plant plumes.

The complexities involved in measuring the numerous parameters necessary to represent the rainout mechanism as it applies to power plant plume depletion have resulted in a large information gap. To realistically model the rainout mechanism requires data on specific cloud parameters such as depth, temperature and water distributions, electrical activity, as well as information about the sulfate aerosol, such as size distribution, concentration, wettability and activity as a nucleus.

While surface contact is probably an important plume depletion process for SO_2, methods of estimating this effect are not sufficiently well developed to make accurate quantitative estimates of a depletion rate. The rate of pollutant removal by vegetation and soil has been shown in laboratory studies to be affected by wind velocity, canopy height, light intensity, soil pH, and moisture content. Based on the laboratory findings, as much as 49 percent of the SO_2 emitted from a large coal-fired plant would be depleted under some conditions within ~32 km due to the average uptake rate of a 40 cm alfalfa canopy during daylight.

Numerical models of the dry deposition process, still rather primitive, have relied mainly on wind tunnel data to predict deposition velocities. They require atmospheric data for validation purposes. The source depletion approach, in which source strength is reduced as a function of downwind distance has been commonly used in Gaussian plume equations to represent the plume depletion processes. A steady-state one dimensional mass transfer model is also available.

PLUME TRANSPORT AND DIFFUSION

Sixteen field observation programs directly related to the diffusion of a plume from a continuous elevated point in complex terrain were reviewed. Most of the field programs have been very limited in scope, either in terms of the intensity of observations or the duration of observations; typically a program consists of one week of plume measurements. The majority of the field programs have focused on observing the worst case situations which have usually been judged to be associated with very stable conditions.

The principal characteristics of flow over complex terrain which create enhanced turbulence are (1) thermally driven drainage and up-slope flows and (2) waves in the lee of a ridge. Channeling and physical diversion of flow may also increase the turbulence.

The most abundant data available to confirm turbulence enhancement due to complex terrain are from bivanes and tetroons. Measurements of plume contaminants confirm the relevance of these measurements to plume behavior. Wind fluctuation data are especially important for estimating plume dispersion rates in rough terrain.

There is some evidence that horizontal spreading (i.e., σ_y), is amplified to a greater extent than vertical spreading in complex terrain, particularly under stable conditions. The resultant increase of σ_y has been attributed primarily to plume separation processes and plume meandering which occur when the higher terrain elevations are confronted by the plume. Overall, the observed plume dimensions in complex terrain (compared to flat terrain for similar stability conditions) have a larger cross-section and the plume is generally more dilute in the core by a factor of two or more.

Although several models are available for estimating ground-level concentrations from an elevated point-source in complex terrain, validation analyses suggest that each is subject to systematic errors which may occur because the effects of terrain are not adequately represented. In general, the models are capable of estimating ground concentrations, measured at distances greater than 10 km from the source, within a factor of two. Larger errors occur for distances closer to the source.

Practical methods of characterizing the effects of terrain on the path and the diffusion of an elevated plume need to be developed. There are many difficulties in providing a working model of mountain wind fields in complex terrain even though adequate theories for local wind in simple ridge and valley situations have been developed. The topography amplifies all the problems typically associated with initializing and grid meshing in mesoscale dynamics modeling. Analytic methods for generating and quantifying terrain characteristics with sufficient reality and economy for modeling are needed.

Results from one series of tracer studies suggest that the turbulent downward transport of the plume during transitional fumigation periods between stable morning and neutral/unstable afternoon conditions, may cause higher ground-level concentrations than the passing of the plume close to or impinging on elevated features under stable conditions. More information (for example, extensive temperature profile data atop ridges near point sources when potential fumigation conditions exist) must be available from field measurement programs to define the behavior mechanisms associated with the highest ground concentrations in complex terrain situations.

Meteorological data used in previous studies, obtained from local radiosondes, pilot balloons, constant-volume balloon flights, aircraft, and surface networks, may not have been an adequate representation of the variation of meteorological conditions throughout the areas of complex terrain which affected behavior of the plumes under study. Readings obtained do not appear to have fully reflected orographically induced eddies, directional wind shear, and thermal convection processes which are vital to the plume transport process. Even with batteries of specially placed sensors, mobile ground vans, and helicopter observations (such as have characterized most of the 16 field studies of plume advection and dispersion in complex terrain examined in this report) it has not so far been possible to assemble and process field data in ways which define the circumstances under which plume impingement (or near impingement) occurs in complex terrain. If this has been the case for data gathered in specially-equipped research programs, an inescapable conclusion is that the kinds of meteorological, air quality, and pollution emissions data now routinely available to air quality analysts for power plant plume studies are inadequate - in the present state-of-the-art plume modeling - for the acceptably accurate calculation of ground-level concentrations due to a power plant plume in complex terrain.

There is a need for the development of improved modeling techniques which will permit the calculation of validatable plume concentration estimates (and plume impingements if these occur) in complex terrain. These modeling techniques should be based upon data which are routinely measured and are routinely available, at an economically affordable cost, for the vicinities of power plant sites.

PART 2

NON-CONSERVATIVE ASPECTS OF PLUME BEHAVIOR

The dispersion and the fate of effluents from power plant stacks are a function of conservative and non-conservative aspects of plume behavior. If the effluent plume were to consist entirely of non-reactive gases it would be transported by wind and diluted by turbulence, but the total mass of the original gas would remain unchanged in the dispersion process. The plume behavior would be conservative. The ideal gas described by conventional Gaussian plume dispersion models has a conservative behavior. In reality such behavior can be approximated for short periods of time by plumes of fine particulates and of relatively non-reactive stack effluents such as sulfur dioxide, but the approximation is inadequate for periods of more than a few hours (or less for more reactive effluents) because of chemical and physical transformations which may change both the composition and the total mass of material remaining in the plume. These chemical and physical transformations are the non-conservative aspects of plume behavior.

Considerations of non-conservative plume behavior must necessarily modify all efforts to provide a realistic, quantitative description of the overall behavior and the ambient air pollution impact of stack plumes from large fossil-fueled power plants - whether in complex or in simple terrain.

A discussion of existing research into non-conservative plume behavior is presented in the next four sections (III, IV, V, and VI) of this report. Conservative plume behavior is discussed subsequently in Part 3.

Section III

PRINCIPLES AND LABORATORY RESEARCH

The combustion of fossil fuels, whether coal, petroleum products or natural gas, is an oxidation process in which carbon and hydrocarbons plus air are converted principally to carbon dioxide and water vapor which are exhausted from the stack as effluents. A residue of solid ash may be removed in the power plant (through the use of precipitators and similar cleaning devices) or the ash may be largely discharged from the stack as particulate matter. Most fossil fuels contain some sulfur, which is largely oxidized to sulfur dioxide and leaves the plant in the stack plume unless in-plant sulfur removal processes are used. Trace metals in the fuels appear in the stack effluents. A by-product of high-temperature fuel combustion is the oxidation of some of the molecular nitrogen in the combustion air to nitric oxide and nitrogen dioxide; these gases also appear in the stack effluent.

Sulfur dioxide is the stack effluent of major interest, because of its known adverse effects upon human health and vegetation, and because of its propensity to oxidize to sulfates which may cause greater damage. All identified pollutants in stack gases are, however, of interest for plume behavior studies.

The main combustion processes of fossil fuels in power plants are accompanied by numerous intermediate side reactions, resulting from the variety of trace chemical constituents, temperatures, and oxygen concentrations encountered within the combustion system. Upon emission, the SO_2 component of the plume is not in chemical equilibrium with its rapidly changing surroundings and enters into further reactions in the plume. Knowledge of the chemistry of sulfur oxides in the atmosphere is still incomplete but, strong evidence indicates that the ultimate fate of sulfur oxides in the atmosphere is oxidation to some form of sulfate, whether by a homogeneous path with reactants in the same phase (the gas phase, liquid phase, or solid phase), or a multi-phased heterogeneous path. The plume-atmosphere interactions which are known to exert an influence on ultimate SO_2 fate are depicted in Figure 1 from Hales et al. (1971a).

It is the intent of this Section III to provide a review of the mechanisms that convert sulfur dioxide to sulfates, the detailed chemistry and dynamic processes associated with these changes, and the

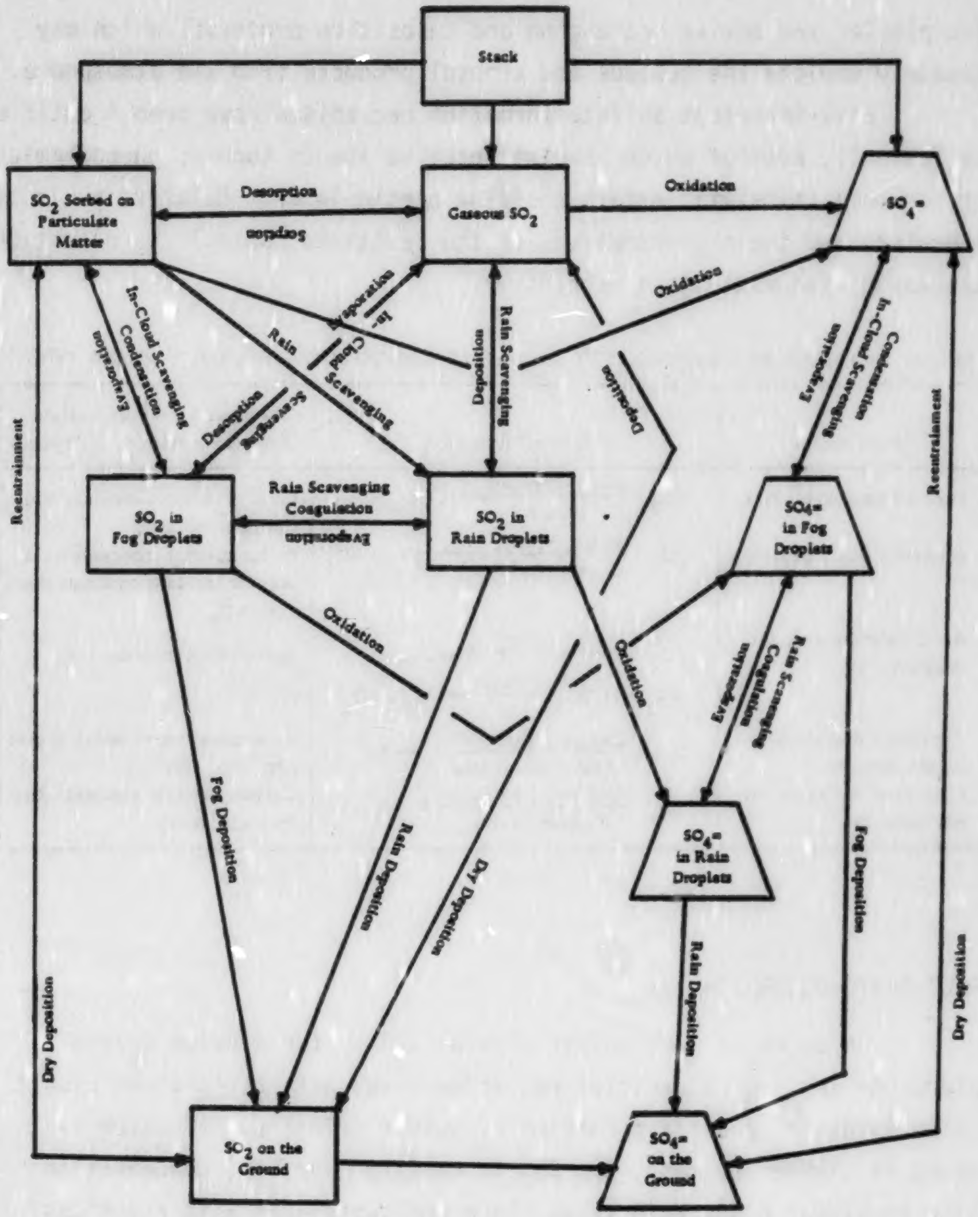
Stack
SO₂ Sorbed on Particulate Matter
Desorption
Sorption
Gaseous SO₂
Oxidation
SO₄=
In-Cloud Scavenging
Condensation
Evaporation
Rain
Scavenging
In-Cloud Scavenging
Evaporation
Desorption
Scavenging
Deposition
Rain Scavenging
Oxidation
Deposition
Condensation
In-Cloud Scavenging
Evaporation
Reentrainment
SO₂ in Fog Droplets
Rain Scavenging
Coagulation
Evaporation
SO₂ in Rain Droplets
SO₄= in Fog Droplets
Reentrainment
Oxidation
Oxidation
Rain Scavenging
Coagulation
Evaporation
SO₄= in Rain Droplets
Fog Deposition
Rain Deposition
Dry Deposition
Fog Deposition
Rain Deposition
Dry Deposition
Dry Deposition
SO₂ on the Ground
SO₄= on the Ground

Figure 1. Possible Atmospheric Paths for SO_2 Upon Emission From a Power Plant Stack (after Hales et al. 1971).

principles of the active scavenging and deposition processes which may ultimately deplete the gaseous and aerosol products from the atmosphere.

Five important sulfate formation mechanisms have been identified (see Table 1), most of which involve reactive agents such as photochemical smog, ammonia, catalytic metals and fine particulates. Relative humidity, temperature and the concentrations of the reactants control the oxidation rates associated with these mechanisms.

TABLE 1. MECHANISMS THAT CONVERT SULFUR DIOXIDE TO SULFATES (from U.S. EPA 1974)

Mechanism	Overall Reaction	Factors on Which Sulfate Formation Primarily Depends
1. Direct Photo-oxidation	$SO_2 \xrightarrow[\text{Water}]{\text{Light, Oxygen}} H_2SO_4$	Sulfur dioxide concentration, sunlight intensity
2. Indirect Photo-oxidation	$SO_2 \xrightarrow[\text{Organic oxidants, OH}]{\text{Smog, Water, NO}_x} H_2SO_4$	Sulfur dioxide concentration, organic oxidant concentration, OH, NO_x
3. Air Oxidation in liquid droplets	$SO_2 \xrightarrow{\text{Liquid water}} H_2SO_3$ $NH_3 + H_2SO_3 \xrightarrow{\text{Oxygen}} NH_4^+ + SO_4^=$	Ammonia concentration
4. Catalyzed oxidation in liquid droplets	$SO_2 \xrightarrow[\text{Heavy metal ions}]{\text{Oxygen, liquid water}} SO_4^=$	Concentration of heavy metal (Fe, Mn) ions
5. Catalyzed oxidation on dry surfaces	$SO_2 \xrightarrow[\text{Carbon, water}]{\text{Oxygen, particulate}} H_2SO_4$	Carbon particle concentration (Surface area)

DIRECT PHOTO-OXIDATION

Exposure of pure sulfur dioxide and sulfur dioxide-oxygen mixtures to certain ultraviolet radiation wavelength regions can result in slow oxidation and the formation of sulfur trioxide. If water is present in the environment, the SO_3 is rapidly hydrated, condensation of the resulting H_2SO_4 acid takes place and subsequent acid reactions with particulate matter are common. Laboratory chamber investigations of the reaction kinetics of homogeneous photo-oxidation are subject to uncontrolled variables and unknown experimental parameters, such as intensity and spectral distribution of radiation, humidity, temperature, gaseous impurities, aerosol impurities, and reactive container surfaces. For example, Cox and Penkett (1970) found the conversion rate for the decay of gaseous SO_2 in natural sunlight was as high as 100 percent per hour and attributed the results to possible wall effects. The interaction of SO_2 with trace impurities in the air was suspected to be the cause of increased condensation nuclei in the dark when SO_2 was activated in the same experiment.

Examination of the absorption spectrum for SO_2 shows two bands above 2900 Å (the second significantly stronger than the first) involved in the transition of SO_2 to an excited state. Early studies sought to examine the quantum yields (ϕ) for the photo-oxidation of SO_2 in the first allowed region and the outcomes of these efforts resulted in a variety of values for a variety of experimental conditions and reactant concentrations. The results with the SO_2 pressures used were generally extrapolated to ppm concentrations usually found in the atmosphere. Of the experiments designed to investigate the photo-oxidation of SO_2 at low concentrations in the air using natural sunlight (see Table 2), conversion rates indicated by the first order rate constants obtained are seen to vary from the low values of 0.05% hr^{-1} - 0.65% hr^{-1} (Hall 1953; Gerhard and Johnstone 1955; Urone et al. 1968; and Cox and Penkett 1970) to that of 24% hr^{-1} (Renzetti and Doyle 1960). The low values are believed the more realistic for low concentrations of SO_2, since most of these workers used very pure gases, resulting in minimal interference from trace impurities. Sethi et al. (1969), Cadle and Allen (1970) and McQuigg and Allen (1970) have presented results which tend to eliminate homogeneous direct photo-oxidation as an important mechanism for oxidation of SO_2 in the troposphere. Their low quantum yield results of $\sim 5 \times 10^{-3}$ and the more recent lower value of 1×10^{-9} reported by Friend et al. (1973) indicate that the mechanism is too slow to account for transformations observed in periods of a few hours. Direct photo-oxidation is possibly important only for extremely long-range transport of low levels of pollution such as on a global scale.

TABLE 2. STUDIES OF THE PHOTO-OXIDATION OF SULFUR DIOXIDE IN SUNLIGHT

Researchers	Laboratory Technique Utilized	Reported Quantum Yield	Conversion Rate Indicated (for Sunlight) By First Order Rate Constant
Hall (1953)	Measured the photo-oxidation rate to SO_3 (56-230 nm) with O_2 (50-200 nm) in sunlight.	10^{-2}	0.05% hr^{-1}
Gerhard and Johnstone (1955)	Measured the amount of sulfuric acid aerosol found in the photolysis of SO_2 at low concentrations in moist air using both a mercury sunlamp (295-305 nm) and natural sunlight.	10^{-3}	0.1 to 0.2% hr^{-1}
Renzetti and Doyle (1960)	Measurements of the decrease in the gas phase SO_2 concentration after SO_2 air mixtures had been exposed to a mercury u. v. lamp for a known time.	3×10^{-1}	24% hr^{-1}
Urone et al. (1968)	Used technique similar to Renzetti and Doyle (1960); compared their 310-420 nm u. v. source with natural sunlight and suggest that lower estimates of photo-oxidation rates in sunlight are more realistic.	-	0.1% hr^{-1}
Cox and Penkett (1970)	Low concentrations of gaseous SO_2 and a sulfur-containing aerosol were monitored both in the dark and sunlight using radioactive $^{35}SO_2$ and the rate of the photochemical reaction determined from SO_2 decay and aerosol yield.	10^{-2}	0.65% hr^{-1}
Friend et al. (1973)	A system was designed for the purpose of studying aerosol formation when trace gases in air are irradiated with ultraviolet light.	1.0×10^{-9}	$<$0.01% hr^{-1}

INDIRECT PHOTO-OXIDATION

Chemical reactions involving SO_2 in mixtures of air containing oxides of nitrogen only, or with oxides of nitrogen and hydrocarbons have a wide range of potential reaction paths. Researchers are seeking to define these paths and the SO_2 oxidation kinetics associated with each of these reaction systems.

Davis (1975b) identified nine possible initiating reactions (see Table 3) of homogeneous gas-phase oxidation processes which might be involved in the conversion of SO_2 to H_2SO_4 aerosol in the troposhere. The lifetimes for conversion range from 10^{11} days down to 3 days for the different reactant species.

TABLE 3. NINE INITIATING REACTIONS FOR HOMOGENEOUS GAS PHASE OXIDATION OF SULFUR DIOXIDE AND THEIR LIFETIMES FOR CONVERSION TO H_2SO_4 IN THE TROPOSPHERE (after Davis 1975b)

Number	Reaction	Estimated Concentration of 2nd Species (Molec/cm^3)	Troposphere Lifetime (Days)
1a 1b	$SO_2 + h\nu(2400\text{-}3400\text{Å}) \rightarrow SO_2^*$ $SO_2^* + O_2 \rightarrow (SO_4)$	---	4×10^7
2	$SO_2 + O(^3P) + M \rightarrow SO_3 + M$	$\sim 1 \times 10^4$	$\sim 6 \times 10^4$
3	$SO_2 + O_2\ (^1\Delta) \rightarrow (SO_4)$	$\sim 10^6$	$\sim 10^8$
4	$SO_2 + O_3 \rightarrow SO_2 + O_2$	1×10^{12}	$\sim 10^5$
5	$SO_2 + NO_3 \rightarrow SO_3 + NO_2$	$\sim 1 \times 10^7$	$\sim 10^{11}$
6	$SO_2 + N_2O_5 \rightarrow SO_3 + N_2O_4$	$\sim 6 \times 10^6$	$\sim 10^9$
7	$SO_2 + HO_2 \rightarrow SO_3 + OH$	$\sim 5 \times 10^8$	~ 23
8	$SO_2 + OH + M \rightarrow HSO_3 + M$	$\sim 5 \times 10^6$	~ 3
9	$SO_2 + CH_3O_2 \rightarrow SO_3 + CH_3O$	$\sim 10^8$	~ 100

In the absence of high organic radical concentrations and catalytic surface sites the prime conversion mechanisms in the atmosphere, as reported by Davis (1975b) and other researchers (Friend et al. 1973, Crutzen 1974), appear to be the SO_2 reaction with either OH or HO_2 (Reactions 7 and 8 in Table 3). Free radicals are the neutral fragments of molecules. In unpolluted tropospheric air, the most important radicals appear to be OH and HO_2 because they are essentially inert toward the main atmospheric constituents (N_2, O_2, Ar, CO_2, and H_2O), but react readily with many trace gases. In particular, the reaction of OH with NO_2 has been suggested as a significant source of atmospheric nitric acid and the reaction of OH with methane has been suggested as a significant (although disputed) source of atmospheric carbon monoxide. The source of the OH radical is the reaction of water vapor with metastable oxygen (O('D)), a species produced by photolysis of ozone. For this reason, the presence of ozone may be indicative of the coexistence of the OH radical (Davis 1976, personal communication). Rate constants must be determined for numerous reactions involving the OH radical to determine the importance of these reactions as efficient conversion mechanisms in the atmosphere. The concentrations of SO_2 and other transient chemical species must be jointly measured to effectively study reactions; a laser-induced fluorescence system has been developed (Lubkin 1975) so that hydroxyl radical concentrations can be analyzed in a more quantitative manner than other currently available techniques permit. Despite the low rates of Reaction 2 at atmospheric concentration levels, this reaction is apparently the most important SO_2 oxidation mechanism in experimental tests of irradiated SO_2-NO_x-air systems. NO_2 added to an SO_2-Air mixture will undergo photolysis and liberate oxygen molecules which encourage Reaction 2. In a study of the NO_2O_5-SO_2-O_3 reaction system, Daubendiek and Calvert (1975) suggest the homogeneous gas-phase reaction systems $NO_3 + SO_2 \rightarrow NO_2 + SO_3$ and $N_2O_5 + SO_2 \rightarrow N_2O_4 + SO_3$ (Reactions 5 and 6) are not important removal paths based on their recent derivation of the rate constants of these reactions.

The addition of hydrocarbons to a system of SO_2 and NO_x and subsequent irradiation results in observed SO_2 oxidation rates that are much greater than in the SO_2-NO_x system. The direct reaction of SO_2 and hydrocarbons has been the subject of study for the past quarter of a century and unfortunately only a few of these studies are available which report a base of experimental data spanning a wide range of initial reactant concentrations to deduce the chemical kinetics of a particular photochemical system. The smog-chamber work of Wilson and Levy (1970 and 1972) and Smith and Urone (1974) are two of the very few studies available which report at least partial concentration-time data.

Smith and Urone (1974) conducted a laboratory experiment utilizing pyrex flasks in which they irradiated 2 ppm [SO_2] and 0, 0.85, 1.7, 3.4, 5.1, and 10.2 ppm [NO_2] respectively in the presence of H_2O. The SO_2 oxidation rate reached a maximum of 3.3 ppm min^{-1} for an initial NO_2 concentration of 0.85 ppm. An initial NO_2 concentration of 10.2 ppm and similar experiment conditions resulted in an observed SO_2 oxidation rate of 1.8 ppm min^{-1}. The initial photochemical rate was 1.7×10^{-4} ppm/min in air with no NO_2 present. The rate was found to increase when the SO_2/NO_2 ratio was 1 or 2, but a decrease was evident when the ratio was 0.6 or less. In experimental conditions duplicating those just described, propane was added to the system in initial concentrations ranging from 3 to 12 ppm. Both sets of data exhibited an inhibition in the SO_2 oxidation rate with increasing NO_2. Hydroperoxyl radicals in the NO_x-Hydrocarbon-SO_2 system may have been responsible for the inhibition effect on the SO_2 oxidation.

Wilson and Levy (1970 and 1972) studied the smog-chamber irradiation of a number of SO_2-NO_x-Hydrocarbon mixtures for hydrocarbon species such as 1-butene, 1-heptene, 2,2,4-trimethylpentane, and toluene. Quantitative values did not come out of these studies because of the high degree of scatter in the experimental data. The rate of SO_2 disappearance was found to correlate with hydrocarbon activity. Relative humidity was found to have a dramatic effect on the overall reaction rate in certain high humidity runs.

Groblicki and Nebel (1971), Cox and Penkett (1972), and McNelis (1974) have investigated the oxidation of SO_2 to H_2SO_4 aerosol when SO_2 was added to olefin-O_3 systems. A (biradical) short-lived intermediate product of the ozone olefin reaction was postulated to react with the SO_2. The consequence of this reaction in plume chemistry can only be speculated upon at this time.

Roberts and Friedlander (1976) irradiated a $96m^3$ teflon bag in natural sunlight to study unfiltered ambient air containing added amounts of SO_2, 1-heptene and NO_x. Other hydrocarbons such as cyclohexene, 2-methyl-2-butene, 2,3-dimethyl-2-butene, and 1,7-octadiene were also studied in less detail. The experiments were designed to study the formation of sulfur containing aerosols under photochemical smog environment conditions. A sulfur balance was carried out on the system by simultaneous measurements of the conversion of SO_2 and the formation of particulate sulfur. The results for all of the experiments were qualitatively similar: SO_2 decay was found to be slight until the O_3 concentration became larger than 5 pphm, at which point there was a sharp downward trend in the SO_2 concentration. Two major behavior regimes were observed in these experiments:

1. As NO is converted to NO_2, the concentrations of SO_2, O_3, 1-heptene and b_{scat} (decrease in visibility) are constant
2. Aerosol formation results from the O_3 induced decay of both SO_2 and 1-heptene.

Depending on the initial SO_2 level (and even with surprisingly low relative humidities), the reaction rates varied from 0 to 99 percent per hour, based on psuedo first order depletion of SO_2. Table 4 summarizes the teflon bag experimental conditions.

TABLE 4. SUMMARY OF TEFLON BAG EXPERIMENTAL CONDITIONS
FOR $SO_2/NO/NO_2/$1-BUTENE SYSTEM
(from Roberts and Friedlander 1976)

	Run Number			
	1	2	3	4
Hydrocarbon	1-Heptene	1-Heptene	1-Heptene	1-Heptene
$[HC]_o$ (ppm)	0.89	1.85	1.66	1.47
$[NO]_o$ (ppm)	0.20	0.19	0.10	0.20
$[NO_2]_o$ (ppm)	0.20	0.10	0.10	0.11
$[SO_2]_o$ (ppb)	0	39	80	86
Δb_{scat} (10^{-4} m^{-1})	2.4	9.5	14.5	14.0
k (% hr^{-1})	0	25-82	21-99	27-72
$[O_3]$ max (pphm)	53	41	33	26
% RH	45	16	14	16
T (° C)	31	35	38	36

In the Roberts and Friedlander study a kinetic scheme was proposed for aerosol formation based on a mechanism similar to that of Cox and Penkett (1972). In the gas phase:

O_3 + Olefin → reactive intermediate
reactive intermediate → condensable organic compounds
reactive intermediate + SO_2 → SO_3 + condensable organic compounds
SO_3 + H_2O → H_2SO_4 + sulfates

What has been determined from all the studies of SO_2 oxidation rates in NO_x-hydrocarbon-SO_2 systems is that known reactions (involving SO_2 for which rate constants have been determined) are in themselves insufficient to account for the oxidation rates in these chemical systems.

AIR OXIDATION IN LIQUID DROPLETS

Important oxidation reactions in the aqueous phase are at least quasi-homogeneous by nature, since all of the participating reactants are in solution. Aerosol particles, which constitute the nucleation sites for droplet formation, are considered as participants in these reactions because molecules of SO_2 and other gases quickly diffuse into the interior of a water droplet where they may directly encounter the nucleating particle and allow the oxidation process to proceed throughout the droplet. This process consists of the hydration and subsequent dissociation of the dissolved gases and oxidation of sulfite or bisulfite ions.

Van den Heuvel and Mason (1963), Scott and Hobbs (1967), McKay (1971), and Miller and DePena (1972) investigated the aqueous phase mechanism of SO_2 oxidation catalyzed by NH_3. Van den Heuvel and Mason (1963) examined the following reaction paths:

$$2SO_2 + O_2 \rightarrow 2SO_3$$
$$SO_3 + H_2O \rightarrow H_2SO_4 \rightarrow 2H^+ + SO_4^=$$
$$NH_3 + H_2O \rightarrow NH_4OH \rightleftarrows NH_4^+ + OH^-$$

The rate of formation of $(NH_4)_2SO_4$ was measured by supporting approximately 100 drops of H_2O on a quartz fiber grid in a 5 liter glass flask exposed to air containing gaseous SO_2 and NH_3. An oxidation conversion rate of 2.5% min^{-1} was obtained by extrapolation of data to conditions in an industrial atmosphere containing 100 μgm^{-3} SO_2 and 10 μgm^{-3} NH_3. The amount of formed sulfate when the sulfur dioxide alone was in air was at least two orders of magnitude smaller than when the sulfur dioxide was mixed with ammonia.

Scott and Hobbs (1967) considered a set of nine chemical equilibrium equations for the SO_2-NH_3-liquid H_2O system, including carbon dioxide equilibria. The rate of production of $SO_4^=$ was assumed to be limited by the oxidation of the sulfite ion described by the first order rate equation

$$\frac{d[SO_4^=]}{dt} = K[SO_3^=]$$

Analysis of the data of Van den Heuvel and Mason (1963) resulted in a choice of K = 0.1 min^{-1} as the rate constant. A plot of the theoretical results obtained from the calculations made with the above assumptions did not result in a limiting value for sulfate formation or a direct proportionality between sulfate and the initial SO_2 partial pressure. The theoretical curves indicated that for the concentration of sulfate in waterdrops as a function of time, the presence of ammonia will increase the amount of sulfate produced by the reaction after it has run for some time. An adequate explanation of the SO_2-NH_3-liquid H_2O system can be based on the theory of this research. The rate of sulfur dioxide oxidation derived in this study was about 2.5 percent hr^{-1}.

McKay (1971) examined the kinetics of sulfate oxidation by taking the rate law of Fuller and Crist (1941) and substituting ionization constants from the Scott and Hobbs (1967) study, suggesting that the reaction is roughly on an order of magnitude faster than Scott and Hobbs had assumed. Evident from the research was the enhancement of the reaction rates with a lowering of the temperatures; the increased solubility of ammonia and sulfur dioxide at lower temperatures are cited as the cause by the authors. At the time of the McKay (1971) research, the large negative temperature correlation in his work had not been noted by other workers in the field, although this feature has now been noted by various other investigators. An oxidation rate of approximately 13% hr^{-1} is suggested from this work.

Miller and De Pena (1972) measured the rate of sulfate ion formation in distilled water drops (raindrop size) for a range of SO_2 partial pressures, following the experimental methodology of Van den Heuvel and Mason (1963), and utilizing the basic model of Scott and Hobbs (1972). The region of experimental values for the first order reaction ($d[SO_4^=]/dt = K [SO_3^=]$) showed $K \approx 3 \times 10^{-3}$ sec^{-1}, in good agreement with the operative value in the study of Scott and Hobbs. Oxidation rates of only 0.1% hr^{-1} were calculated from the Miller and De Pena data.

METALLIC-CATALYZED OXIDATION IN LIQUID DROPLETS

Sulfur dioxide may be oxidized to H_2SO_4 in aqueous solution according to the following process:

$$2SO_2 + 2H_2O + O_2 \xrightarrow{catalyst} 2H_2SO_4$$

which requires water, dissolved oxygen and the presence of a catalyst. Ammonia has already been previously mentioned as one such catalyst; metal ions such as Fe^{3+}, Mn^{2+}, and Cu^{2+} constitute the other major grouping of catalyst-oxidation candidates in the atmosphere (Johnstone and Coughanowr 1958). These metal ions are contained in iron and manganese salts normally found as suspended particulate matter in the ambient air from burned coal fly ash. The particulates behave as sites of nucleation which at higher humidities may either dissolve in the droplets or remain as solid condensation nuclei centers. The role of iron as an efficient catalyst in the oxidation of SO_2 in the atmosphere has been studied by Junge and Ryan (1958), Johnstone and Coughanowr (1958), Foster (1969), Brimblecombe and Spedding (1974) and Freiberg (1974 and 1975).

Junge and Ryan (1958) examined the iron-catalyzed reaction of SO_2 oxidation in acid solutions, without investigation of the kinetics. Air containing SO_2 was bubbled through dilute catalyst solutions and a linear relationship was found between the final extent of $SO_4^=$ formation and the SO_2 partial pressure of the reaction. The pH of the catalyst solutions was cited as the governing factor of the SO_2 oxidation process; SO_2 solubility decreases as acidity is increased due to sulfate formation, and eventually a situation is attained where sulfate formation ceases because of liquid phase SO_2 depletion. This bubbling technique has been criticized by Cheng et al. (1971) as an unrealistic representation of an aerosol gas system due to the influences of mixing effects and mass transfer mechanisms on the kinetics of the reaction.

Johnstone and Coughanowr (1958) approached the absorption and reaction aspects of their catalytic oxidation studies by mathematically considering the reaction system as one of gas diffusion into a liquid drop with a concurrent zero order reaction in the liquid phase. The value of the zero order rate constant was acquired from previous experiments involving homogeneous SO_2 liquid phase oxidation. In an experiment which exposed a single drop of manganese sulfate solution to an environment consisting of air and trace amounts of SO_2, it was estimated that manganese sulfate crystals of 1 micron at 1 ppm SO_2 yielded a rate of oxidation in fog droplets on the order of 1 percent min^{-1}. Cheng et al. (1971) had doubts about the validity of the assumption that the reaction rate was controlled by liquid phase gas diffusion.

Foster (1969) derived theoretical growth rates of H_2SO_4 droplets to test the effectiveness of $MnSO_4$ crystallites as a catalyst for SO_2 oxidation in humid, polluted surroundings. Review and comparison with previous theoretical works on the subjects of manganese and iron solution catalytic oxidation of SO_2 led to the following rate expressions, the symbols of which are explained in Table 5, along with the values (when provided) utilized for the calculations:

$$\begin{array}{c}\text{Rate of } SO_2 \text{ oxidation} \\ \text{by } Mn^{+2} \text{ catalyst}\end{array} = \frac{22.4}{10^{-6}} \; \frac{K_i C_i^2 V}{GD} \times 100\% \ \text{min}^{-1}$$

$$\begin{array}{c}\text{Rate of } SO_2 \text{ oxidation} \\ \text{by } Fe^{+2} \text{ catalyst}\end{array} = 22.4 \; \frac{DWK_H \, (1 - fo)}{[H^+]} \; \frac{K_i n_i f_i}{M_i} \times 100\% \ \text{min}^{-1}$$

When these values are substituted into the equations, SO_2 plume oxidation rates of 0.09% min^{-1} for Mn and rates of 0.15 - 1.5% min^{-1} for Fe are the results. This work implies that the major catalytic role for aqueous phase SO_2 oxidation in plumes is played by oxides of iron.

TABLE 5. NOMENCLATURE AND VALUES USED FOR PLUME SO_2 OXIDATION CALCULATIONS (from Foster 1969)

		Nomenclature	Value
General	W	Effluent dust burden, g/liter	2×10^{-3}
	G	Effluent SO_2 content, ppm	$2{\cdot}5 \times 10^{3}$
	D	Effluent dilution factor	10^{-3}
	f_o	Fraction of total sulfur oxidized	10^{-1}
	S	Droplet sulfate concentrations, mol/liter$_{H_2O}$	1
Manganese (Mn_3O_4)	M_i	Oxide molecular weight, g/mol	$2{\cdot}29 \times 10^{2}$
	n_i	Number of catalytic ions per molecule	3
	f_i	Fraction by weight of dust soluble	2×10^{-4}
	K_i	Rate constant	*
	C_i	Catalytic concentration within the droplets	*
Iron (Fe_2O_3)	M_i	Oxide molecular weight, g/mol	$1{\cdot}60 \times 10^{2}$
	n_i	Number of catalytic ions per molecule	2
	f_i	Fraction by weight of dust soluble	10^{-2}
	K_H	Solubility constant	*

* Values not provided.

Matteson et al. (1969) studied the mechanism of SO_2 oxidation catalyzed by manganese sulfate aerosol. The kinetic theory proposed for the oxidation process was based on a four-step chemical reaction involving the formation of intermediate complexes.

$$Mn^{2+} + SO_2 \rightleftarrows Mn{\cdot}SO_2^{\;2+}$$
$$2Mn{\cdot}SO_2^{\;2+} + O_2 \rightleftarrows [(Mn{\cdot}SO_2^{\;2+})_2{\cdot}\ O_2] \rightleftarrows 2Mn{\cdot}SO_3^{\;2+}$$
$$Mn{\cdot}SO_3^{\;2+} + H_2O \rightleftarrows Mn^{2+} + HSO_4^-$$
$$HSO_4^- + H^+ \rightleftarrows H_2SO_4.$$

The rate expression which was determined was

$$-\frac{d(SO_2)}{dt} = K[Mn^{2+}]_0^2$$

where the experimental rate constant = 2.4×10^5 mole^{-1} s^{-1}.

Negligible $SO_4^=$ concentrations were formed for relative humidities below 95 percent in the experiments apparently as a result of inadequate hydration of the metal salt. Rates on the order of 0.5% day^{-1} were recorded. The experimental data obtained were basically incorrect (Cheng et al. 1971) due to errors in the recorded reaction times.

Selected metal salts which had been reported to act as aerosol catalysts ($MnSO_4$, $MnCl_2$ and $CuSO_4$) were tested for their effectiveness in promoting atmospheric oxidation of SO_2 to H_2SO_4 in laboratory work by Cheng et al. (1971). The experiment was carried out using an aerosol stabilizing technique, whereby teflon beads with deposited aerosol particles were packed into a flow reactor, and exposed to influent SO_2 concentrations ranging from 3 to 18 ppm in humid air. Higher SO_2 oxidation rates were found to be positively correlated with higher relative humidities; in addition, the chemical reaction in the liquid phase in conjunction with absorption of SO_2 by aqueous catalyst drops controlled the overall rate of reaction. Using NaCl as a base for comparison, aerosols of $MnSO_4$, $MnCl_2$, and $CuSO_4$ were found to be 12.3, 3.5, and 2.4 times more effective than NaCl in assisting the oxidation of SO_2 on a milligram-to-milligram basis. To approximate the rate of absorption and oxidation of SO_2 by fog droplets in a natural fog, laboratory results were extrapolated to conditions typical of an urban industrial atmospheric environment. The following assumptions were made for this estimation:

1. 15 µm was the average diameter of fog droplets
2. SO_2 concentration in the atmosphere was 0.1 ppm.
3. Fog droplet concentration of catalyst is equivalent to 500 ppm $MnSO_4$; one-half of the fog droplets contain the catalyst capable of oxidizing SO_2 to H_2SO_4
4. 0.2 g of water per cubic meter of air defined the fog concentrations.

The atmospheric oxidation rate under these conditions was found to be 2 percent hr^{-1}.

Brimblecombe and Spedding (1974) noted that many of the experimental data used in the interpretation of the oxidation of SO_2 within the atmosphere in previous studies were characterized by concentrations several orders of magnitude higher than those expected in the aqueous aerosol, and were therefore unrealistic. These researchers attempted to establish the rate of oxidation of low concentrations of SO_2 (about 10^{-5} molar) in aqueous solutions containing traces of Fe^{3+} (about 10^{-6} molar) as these concentrations approach those which might be likely in the atmospheric aerosol. A radiochemical method utilizing $^{35}SO_2$ monitored the oxidation process in the experimental procedure over a range of added Fe^{3+} catalyst concentrations up to 1.6×10^{-8} molar. A radical mechanism was favored due to the high reaction rates noted in dilute solution; hydroxylated Fe^{3+} was postulated as the initiator in the production of $SO_3^=$ radicals:

$$SO_3^{2-} + FeOOH + 3H^+ \rightarrow Fe^{2+} + 2H_2O + SO_3^-$$

To explain the termination of the radical chain, Brimblecombe and Spedding (1974) state that the Fe^{2+} is oxidized back to the +3 oxidation state by reacting as follows:

$$SO_5^- + Fe^{2+} \rightarrow SO_5^{2-} + Fe^{3+}$$

Small amounts of Fe^{3+} in the atmosphere would be capable of oxidizing a large quantity of sulfur dioxide by such a step which would allow regeneration of Fe^{3+}: $2Fe^{3+} + H_2O + H_2SO_3 \rightarrow SO_4^{2-} + 4H^+ + 2Fe^{2+}$. Assuming an SO_2 concentration of 28 $\mu g/m^3$ and a Fe^{3+} concentration of 10^{-6} molar, Brimblecombe and Spedding arrive at a SO_2 conversion rate in fog of ~ 3.2 percent/day.

Freiberg (1974, 1975) examined the mechanism of iron-catalyzed oxidation of SO_2 in oxygenated acid solutions by beginning with the rate expression:

$$\frac{d[SO_4^=]}{dt} = \frac{K_c K_d^2\ [H_2SO_3]^2\ [Fe^{3+}]}{[H^+]^3}$$

where

K_c = rate constant

k_d = first dissociation constant of sulfurous acid

Since the pH increases as relative humidity rises, the reaction rates were found to be especially dependent on high relative humidity; an increase from 80 percent to 90 percent RH resulted in an eightfold increase of iron-catalyzed oxidation of SO_2, whereas a relative humidity increase from 80 percent to 95 percent resulted in a sixty-four-fold increase in the oxidation reaction rate. Freiberg also examined the dependence of the oxidation rate of SO_2 on temperature. The relationship involves numerous physical-chemical processes which influence the reaction in a way different from the expected reaction rate increase with temperature alone; the end result is a significant decrease in the yield of the reaction as the temperature increases. An increment of 5°C results in a net decrease in reaction rate of iron catalyzed oxidation of SO_2 by about an order of magnitude.

OTHER AQUEOUS SO_2 OXIDATION MECHANISMS

When water droplets are present in a mixture of SO_2, O_3 and air, both SO_2 and O_3 are observed to vanish swiftly, indicating that the reaction $SO_2 + O_3 \rightarrow SO_3 + O_2$ proceeds more efficiently in the liquid phase than it does in the gas phase.

The mechanism of aqueous phase SO_2 oxidation by ozone has been investigated by Espenson and Taube (1965), Penkett (1972), and Penkett and Garland (1974). In the study by Espenson and Taube (1965), two elements of this mechanism were examined; namely the acid solution reaction of $SO_2 + O_3 + H_2O \rightarrow H_2SO_4 + H^+ + O_2$ and the basic solution reaction of $SO_3^= + O_3 \rightarrow SO_4^= + O_2$. Unfortunately, quantitative information did not result from the work.

Penkett (1972) investigated the reaction path of $HSO_3^- + O_3 \rightarrow HSO_4^- + O_2$ (SO_2 in the form of bisulfite ions) with the rate of oxidation provided by $d[O_3]/dt = K[O_3]\ [HSO_3^-]$. In the kinetic work, the rate constant K = 3.32 ± 0.13 x 10^5 $M^{-1}s^{-1}$, and a 0.05 ppm ozone concentration resulted in an extrapolated SO_2 oxidation rate of ≈ 0.21% min^{-1} at 9.6°C; this compares to the slower rate of 2.5% hr^{-1} predicted by Scott and Hobbs (1967) for SO_2 oxidation catalyzed by NH_3.

Penkett and Garland (1974) studied the same reaction path as Penkett (1972) and, for a pH range from 4 to 7 at 10°C with 0.1 ppm SO_2 and 0.05 ppm O_3 in fog water, determined the reaction rate coefficient of the expression $-d[SO_3^=]/dt = K[SO_3^=]$ to be $K = 4.18 \times 10^{-4} + 1.77\,[H^+]^{1/2}\ sec^{-1}$.

HETEROGENEOUS STUDIES

Corn and Cheng (1972) examined the heterogeneous catalysis of sulfur dioxide in the laboratory with respect to interactions with insoluble particulate matter. Micron and submicron particles of $CaCO_3$, V_2O_5, Fe_2O_3 and flyash from a coal-burning power plant were utilized along with suspended particulate matter from an urban environment, MnO_2 and activated carbon. Through the use of an aerosol stabilizing technique, the aerosol particles were deposited on teflon beads which were packed into a flow reactor in order to obtain kinetic data. MnO_2, Fe_2O_3 and the suspended particulate matter from Pittsburgh air were found to sorb SO_2, with physical adsorption as the primary process. Steady-state rates of conversion of 0.013 and 0.021 μg of SO_2 min^{-1} mg^{-1} at SO_2 concentrations of 8.0 and 14.4 ppm, respectively, were found for the activated charcoal. Under the conditions of the tests, $CaCO_3$, V_2O_5 and flyash were discovered to be essentially inert to SO_2 at room temperature. Twenty mg of V_2O_5 at 95 percent relative humidity removed only 1.2 percent of the total SO_2 after 50 minutes of exposure, which is surprising, considering that at a high temperature of 500°C or beyond, vanadium pentoxide is known to participate in the oxidation of SO_2 to SO_3 according to the reaction:

$$V_2O_5 + SO_2 \rightarrow V_2O_4 + SO_3$$

Low et al. (1971) studied the gas-solid reaction of SO_2 with CaO by means of infrared spectroscopic techniques as a segment of research designed to investigate the removal of SO_2 from power-plant stack gases by way of catalytic oxidation of powdered limestone introduced into the stack stream. A similar study was conducted by Goodsel et al (1972) for sorption of SO_2 on MgO in research to find the removal rates of SO_2 from plant stack gases by catalytic oxidation with introduced dolomite. Both studies showed the formation of a surface sulfite as the main irreversible interaction product when the CaO and MgO were degassed. Sulfates were formed when the surface sulfite was exposed to heat in an oxygen environment.

A combination of colorimetric and radio-tracer techniques were employed by Urone et al. (1968) in studies of reactions of parts per million concentrations of sulfur dioxide, with laboratory environments containing hydrocarbons, nitrogen dioxide, moisture, particulates and ultraviolet radiation. Powdered oxides of aluminum, calcium, chromium, iron, lead and vanadium were found to react with SO_2 within minutes without the presence of sunlight or ultraviolet radiation, whereas gaseous mixtures of SO_2 and clean air kept in the dark for days did not react. A reaction rate on the order of 0.1 percent hr^{-1} was found for the photochemical reaction of sulfur dioxide in clean air with ultraviolet radiation equivalent to noonday sunlight. Inert solids such as NaCl and calcium carbonate resulted in negligible heterogeneous reaction rates with SO_2; however the reaction was significant for heterogeneous reactions involving other particulate matter such as iron and lead in some instances, as shown in Table 6.

TABLE 6. SUMMARY OF EIGHT EXPERIMENTS INVOLVING SO_2
REACTIONS IN THE PRESENCE OF VARIOUS PARTICULATES
(from Urone et al. 1968)

Original SO_2 Concentration	2nd Reactant	2nd Reactant Concentration	UV Exposure	Total Time in Flask	SO_2 Reaction Rate
14 (ppm)	$CaCO_3$	30.3 (mg)	0 (min)	1100 (min)	0.2% hr^{-1}
14	CrO_3	11.0	0	1030	0.5% hr^{-1}
8	V_2O_5	17.2	180	810	0.7% hr^{-1}
17	PbO	11.0	0	15	1.7% min^{-1}
18	PbO_2	12.0	0	9	6.0% min^{-1}
10	($CaO + A\ell_2O_3$)	(16.6, 19.8)	(0, 0)	30	3.0% min^{-1}
14	$A\ell_2O_3$	33.1	0	1145	.04% min^{-1}
8	Fe_3O_4	14.0	0	4	17% of SO_2 reacted

Smith et al. (1969) developed a method for studying adsorption at solid-gas interfaces in aerocolloidal systems by generating submicron sized aerosols (0.01 to 0.1 μm) using an exploding-wire technique and radio-labeled-gases to achieve adequate sensitivity in sorption measurements. Preliminary measurements of ^{35}S-labeled SO_2 with Fe_3O_4, Al_2O_3, and platinum aerosols at ambient conditions, showed that low SO_2 concentrations resulted in preferential chemisorption followed by multilayered physical adsorption at higher concentrations. Although significant adsorption was evident on the Fe_3O_4, Al_2O_3 and platinum surfaces, the oxidation rates were not presented in the Smith et al. (1969) research. Overall, it is seen that dry heterogeneously catalyzed SO_2 oxidation systems are relatively ineffective when compared to aqueous phase oxidation systems.

The sulfate forms that result as products of SO_2 oxidation in the atmosphere are tabulated in Table 7 along with brief descriptions of their chemical nature (Wilson et al. 1976). The sulfate types are secondary in nature; primary surface types (e.g., those emitted directly by industrial or by natural sources) are not listed. The probable size class by mass particle diameter for each of these sulfate forms is between 0.1 and 1.0 μm. Whitby and Liv (1974) examined the size distribution of the general atmosphere aerosol, and found the number concentration to be dominated by particles in the 0.002 to 0.02 μm range and the major part of the aerosol surface area in the 0.05 to 0.5 μm size range; a bimodal distribution characterized particle volume and mass. The "accumulation mode," defined as the size range between 0.1 and 1.0 μm, is estimated to be the size fit for 80 percent of atmospheric sulfate. Secondary accumulation-mode aerosol can have a very long life time in the atmosphere, traveling great distances, and having the opportunity to participate in other chemical processes before removal by fallout or rainout.

TABLE 7. A SUMMARY OF TYPICAL SECONDARY ATMOSPHERIC SULFATE TYPES
(from Wilson et al. 1976)

Formula	Names	Sources	Notable Chemical Properties
H_2SO_4	Sulfuric acid (oil of vitriol)	Atmospheric oxidation of SO_2; direct from manufacturing.	Strong acid, very hygroscopic (drying agent at low RH).
NH_4HSO_4	Acid ammonium sulfate	Oxidation of SO_2 with NH_3 addition.	Strong acid, hygroscopic.
$(NH_4)_3H(SO_4)_2$	Triammonium acid disulfate (Letovicite)	Oxidation of SO_2 plus NH_3.	Acidic, deliquescent at ~ 65 percent RH.
$(NH_4)_2SO_4$	Ammonium sulfate (Mascagnite)	Oxidation of SO_2 plus NH_3.	Weak acid, water soluble deliquescent at 80 percent RH.

Charlson et al. (1974) investigated the hygroscopic properties of the sulfate aerosol in the St. Louis region and found that the H_2SO_4 can absorb water and increase in size at all humidities, as can NH_4HSO_4; while $(NH_4)_2SO_4$ can do so at relative humidities above about 65 percent. Sulfuric acid is known to give rise to heteromolecular nucleation of droplets in the presence of water vapor, and will condense on preexisting nuclei (Stauffer et al. 1973 and Cox 1973). Unfortunately, detailed estimates of growth parameters for aqueous solution droplets of sulfuric acid are not available at this time. Research on the growth of solution droplets of sulfuric acid as well as other important solutes as a function of relative humidity will be useful in the analysis of aerosol growth curves to identify the chemical composition and understanding of the solute-solvent interaction in the process of growth of solution droplets. Nair and Vohra (1975) indicate that the relative increases in size and mass depend on the quantity of solute per particle. They show that sulfuric acid nuclei in the Aitken size range (< 0.1 μm) can grow to cloud nuclei as the relative humidity increases. Nuclei can grow continuously at relative humidities below 100 percent due to the instability produced by the chain process of sequential condensation of water and solute molecules onto the nuclei. Katz and Mirabel (1974) examined how certain kinds of suspended liquid aerosols can be generated in the atmosphere by the binary homogeneous nucleation of water vapor and a polluting reactant such as sulfuric or nitric acid; humidities as low as 10 percent in the presence of H_2SO_4 were found to result in large nucleation rates.

RELATED CHEMICAL PROCESSES

Field programs designed to investigate the plume chemistry of processes other than sulfate formation are rare in the literature. An excellent example of one such study is the Davis et al. (1974) investigation of nitrogen oxides and SO_2 chemistry in the Morgantown power plant plume, in which the power plant plume was postulated to have had a significant effect in producing increased ambient O_3 concentrations far downwind of the plant. The Morgantown power plant plume investigation is described in Section V.

Whereas sulfur dioxide plume chemistry primarily involves the transformation of SO_2 to sulfate ($SO_4^{=}$), nitrogen oxide chemistry involves the transformation of NO to NO_2, with the subsequent formation of ozone, other oxidants, and nitrates. The literature is filled with photochemical reactions involving O_3, NO, NO_2, hydrocarbons and free radicals. These data have been a product of laboratory work concerned with the polluted urban environment, or more recently have been data gathered in studies of rural ozone and in attempts to determine the downflux of stratospheric ozone.

The photolysis of nitrogen dioxide is the only reaction known to generate important numbers of oxygen atoms necessary for the production of ozone:

$$NO_2 + h\nu \rightarrow NO + O \qquad (1)$$

Once the oxygen atoms are present, they react with oxygen molecules, involving a nitrogen molecule or another oxygen molecule as the third body to form ozone according to:

$$O_2 + O + M \rightarrow O_3 + M \qquad (2)$$

Reaction (1), the photolysis of NO_2, is a very rapid reaction, but simultaneously, the reaction of ozone with NO is a rapid O_3 removal mechanism:

$$NO + O_3 \rightarrow NO_2 + O_2 \qquad (3)$$

and additionally, ozone is also removed by a relatively slow reaction with NO_2:

$$NO_2 + O_3 \rightarrow NO_3 + O_2 \qquad (4)$$

Chemical reactions involving organic molecules or radicals which drive or maintain the concentration ratio of NO_2 to NO at high levels are responsible for high steady-state (i.e., O_3 production rate = O_3 destruction rate) levels of ozone. In a polluted urban environment, depending on the nature of the hydrocarbons present (i.e., paraffin, olefin or aromatic), a branching reaction sequence occurs which allows photolysis of NO_2 to create additional NO_2 at the expense of NO. Figure 2 from Hanst and Bufalini (1971) illustrates how various hydrocarbons will oxidize NO at different rates in smog-chamber experiments. It has been theorized that for power plant plumes, a mechanism not affiliated with hydrocarbons could oxidize NO + NO_2 to elevate the $[NO_2]/[NO]$ value, thus increasing ozone concentrations. Two such mechanisms have been suggested; the first, a sulfur mechanism and the second, a chlorine mechanism (Tesche et al. 1976); however recent studies of plumes in areas with low background hydrocarbon concentrations have detected no net ozone production.

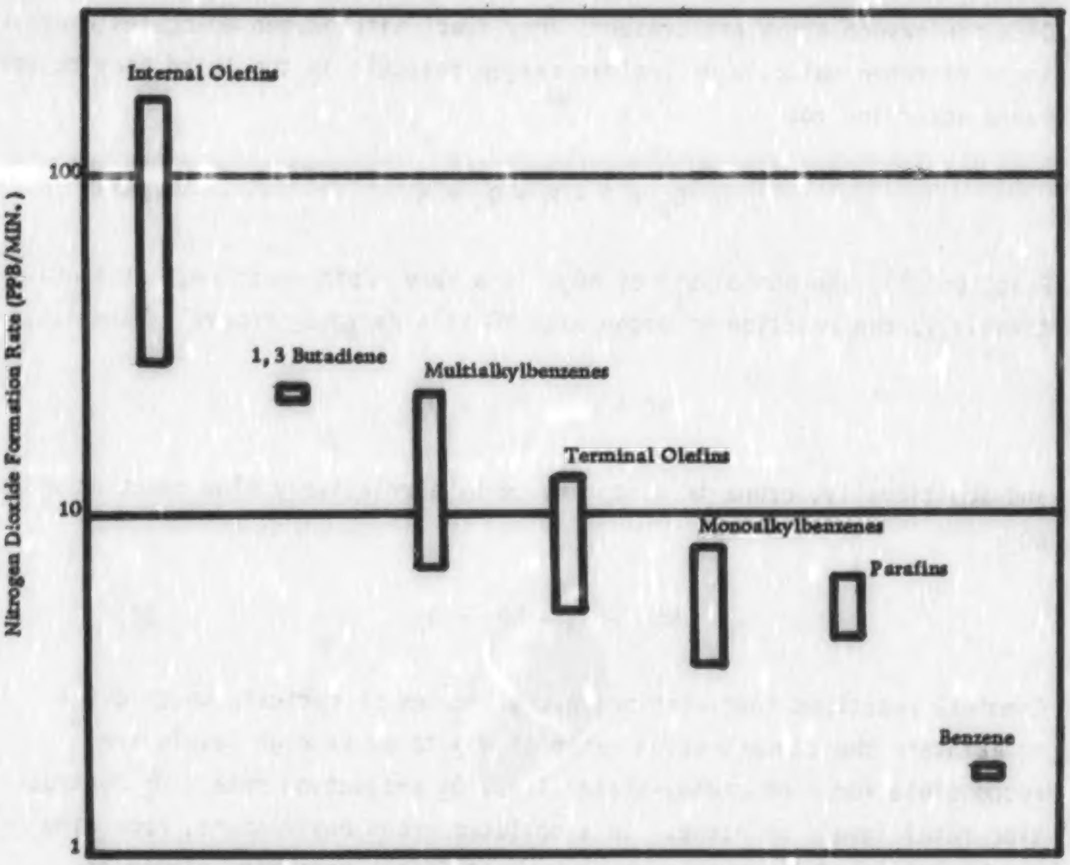

Figure 2. Effect of Hydrocarbon Structure on Nitrogen Dioxide Formation Rate (From Hanst and Bulfalini 1971).

PRECIPITATION SCAVENGING

Two basic processes act to scavenge an aerosol from the atmosphere. The first process is the result of the aerosol serving as a cloud nucleus or undergoing capture by cloud water or ice particles, and is termed "rainout." The removal of the subcloud aerosol by raindrops as they fall is the second process, "washout."

Fundamentals of the influence of rain on the precipitation scavenging of gases have been reviewed by Postma (1970) and Hales (1972). The possibility that a gas plume could become redistributed in the atmosphere due to the action of "reversible" washout has been discussed by Hales. The gas concentration in each raindrop, as it traverses a plume, approaches a concentration saturated with respect to the concentration of the plume (Slinn 1974). The drop can emerge from beneath the plume with a concentration supersaturated with respect to ambient conditions, if no irreversible chemical conversion of the gas occurs within the drop, and desorption of the gas is possible. An increased dosage to low-level receptors is the effective result of this lowering of the plume centerline elevation. A complete mathematical description of the rate at which this sorption-desorption phenomenon might lower a gas plume is a formidable undertaking unless a number of assumptions are imposed. Slinn (1974) assumed it was reasonable to use a convective diffusion equation (assuming the stack exit is high enough and the atmosphere stable enough), ignoring longitudinal diffusion compared with transport, neglecting wind shear, and treating the diffusion coefficients as constants in his attempt to predict this rate of plume redistribution. The "washdown" speed of the plume is a function of the rainfall rate and Henry's law constant (dimensionless). An example cited shows that an SO_2 plume exposed to a rainfall rate of 10^{-4} cm s^{-1}, with a Henry's law constant of 3.3×10^3, results in a velocity of 3.33×10^{-1} cm s^{-1}. Another example demonstrated that a plume would have fallen 30 m after a time of 10^4 s.

Bolin et al. (1974) described the efficiency of the rainout mechanism as dependent on (a) the efficiency of the microphysical processes in the clouds and (b) the frequency of precipitation. The consumption of aerosol particles by the cloud drop varies greatly with particle radius; and the absorption of gases to cloud elements depends on the chemical composition of the cloud drops. More work is required to understand these processes quantitatively but it is interesting to note that there is evidence of a rather rapid transformation of sulfur dioxide into sulfuric acid in clouds as long as the pH of the cloud drop water is significantly higher than 4 (Brosset 1973). It is clear, however, that the most important factor in determining the efficiency of the rainout mechanism for removal of pollutants from the atmosphere is the frequency of rains (Rodhe and Grandell 1973). If rainout were the only sink mechanism, the average residence time for a pollutant in the atmosphere estimated in the Rodhe and Grandell Swedish study would be about 40 hours in the winter and 90 hours in the summer based on seasonal precipitation data collected during a one-year period. Low intensity precipitation periods were appreciably longer in winter than in summer. Rodhe and Grandell also derived the distribution function for the probability of rainout of a pollutant released at an arbitrary instant.

Hales et al. (1971) describe the complex processes of liquid mixing in a falling drop and their relation to mass-transfer, obscured further by film effects at the interface and by the possibility of chemical reaction within the liquid (see Figure 3). Hales (1972) explains that in order for pollutant gas molecules to be captured, they must first migrate from the atmosphere to the surface of the liquid. They then may pass through the liquid-vapor interface and, finally, migrate into the liquid interior where they may react chemically or exist simply as unreacted dissolved gas molecules. The existence of dissolved gas molecules results in a concentration of gas in the liquid which, owing to the finite solubility of the gas, will be characterized by a vapor pressure. Thus, there will be a tendency for gas molecules to desorb from the liquid and, retracing the mechanisms by which they were captured, return to the atmosphere. The general problem of modeling microphysical systems in the atmosphere is discussed in Section IV as it relates to the pertinent microphysical phenomena of drop-gas interactions.

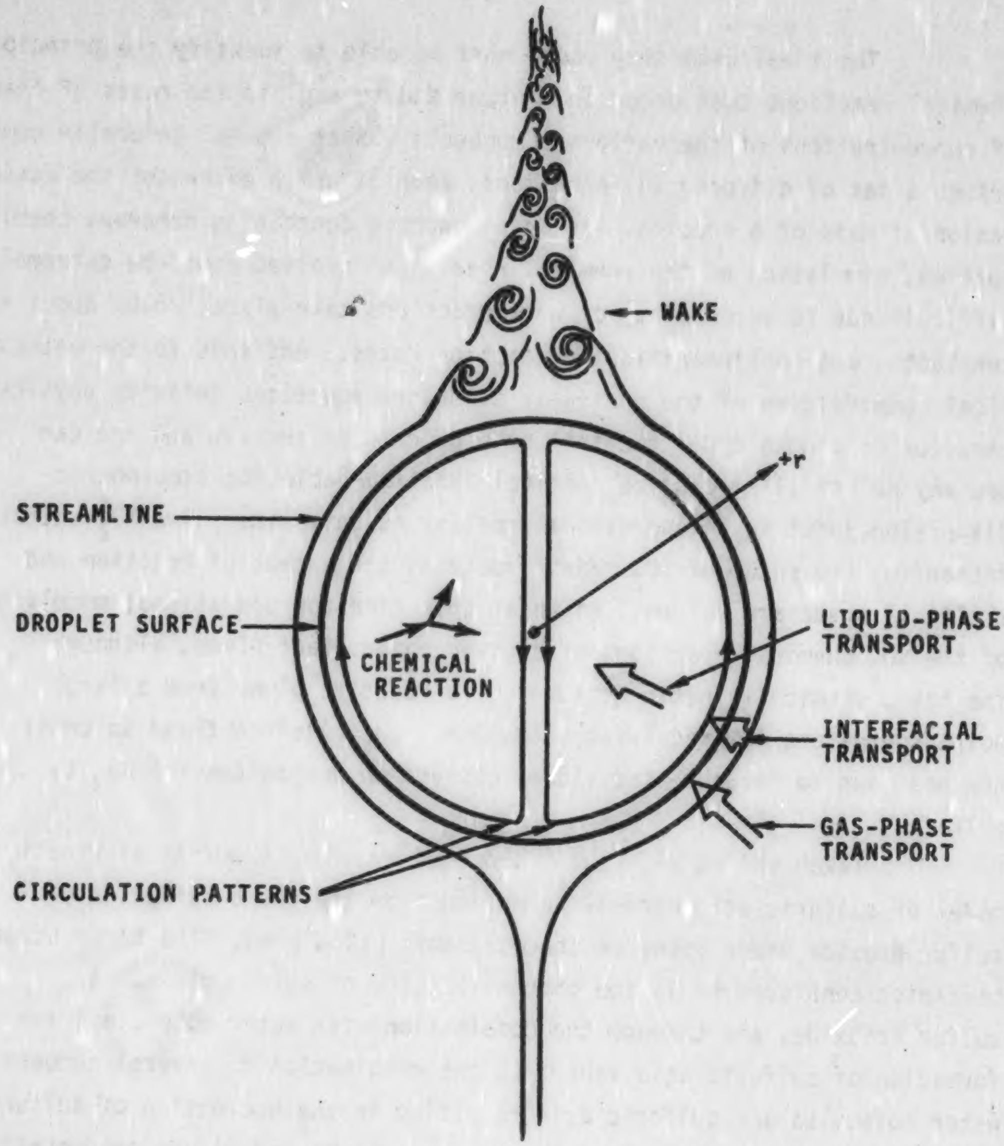
WAKE
+r
STREAMLINE
DROPLET SURFACE
CHEMICAL REACTION
LIQUID-PHASE TRANSPORT
INTERFACIAL TRANSPORT
GAS-PHASE TRANSPORT
CIRCULATION PATTERNS

Figure 3. A Falling Raindrop and the Associated Transport and Washout Mechanisms Acting Upon it in the Atmosphere.

Section IV

MODELS OF PLUME CHEMISTRY AND DEPLETION-DEPOSITION PROCESSES

The ideal chemistry model must be able to identify the principal chemical reactions that occur in a plume and to explain the rates of change of concentrations of the various components. Such a model generally comprises a set of differential equations, each of which expresses the conservation of mass of a species. In an atmosphere containing numerous chemical species, simulation of the chemical reactions involved could be extremely difficult due to uncertainty of what reactions take place, doubt about rate constants, and nonlinearities in reaction rates. Add this to the mathematical complexities of the nonlinear transport equations defining physical behavior of plumes under the influence of complex terrain and one can see why no one all-inclusive chemical reaction-depletion atmospheric dispersion model is an operational reality at this time. The SO_2-sulfate mechanism, the rates of SO_2 transformation, the extent of reaction and the final state are not well known at this time for operational modeling of the SO_2 chemical reactions of a given power plant plume, although the basic chemical kinetic features of a reactive plume from a large point source are now receiving attention. The research trend up until now has been to focus on individual conversion mechanisms for SO_2 to sulfate.

Takahashi et al. (1975) proposed a purely theoretical kinetic model of sulfuric acid aerosol formation from the photo-oxidation of sulfur dioxide vapor based on thermodynamic principles. The three stage mechanism consisted of 1) the photo-oxidation of sulfur dioxide to sulfur trioxide, and through the combination with water molecules, the formation of sulfuric acid vapor; 2) the combination of several numbers of water molecules and sulfuric acid resulting in the nucleation of sulfuric acid vapor to a critical sized cluster or embryo; and 3) the condensation of sulfuric acid and water molecules, as well as through the coagulation with other particles to allow growth of the nucleated embryo to a larger aerosol particle. The nucleation rate for a system of water and sulfuric acid vapor was calculated at various relative humidities according to the expression:

$$I = C \exp(-\Delta G/RT)$$

where

C is a factor containing the probability of capture of a molecule of vapor by a critical embryo
ΔG is the free energy to form an embryo
R is the gas constant
T is the absolute temperature

The nucleation rate was found to increase rapidly with an increase of sulfuric acid vapor pressure, and was relative humidity dependent. Equations were formulated for the number concentration of sulfuric acid vapor and particles, the time change of the number of sulfuric acid molecules in a particle, the deposition rate of sulfuric acid vapor to already formed aerosol particles, and the equilibrium particle radius. Generally the sulfuric acid vapor molecule concentration was found to increase rapidly at the initiation of irradiation and then slowly decrease after reaching a maximum value, determined by the photo-oxidation rate. Indications were that nucleation was predominant in the early stage of the process, with vapor condensation becoming significant in the later stage. In the early stages when nucleation was predominant, the vapor concentration was not always high enough for effective condensation to occur, leading to growth in particle size. However, in some cases of low relative humidity, the nucleation rate was very small so that most of the sulfuric acid vapor condensed to make the particles grow larger. Thus, the relative humidity and the acid vapor oxidation rate characterized the rate of growth of particles. Although its time change is very slow, the sulfuric acid concentration in particles increased, as the condensation of vapor became effective. Unpublished experimental data by Cox (1973) and Takahashi et al. (1975) indicated that particle number concentration at a relative humidity as low as 10 percent or less was much higher than that predicted by this kinetic model; differences were partially attributed to uncertainity in the calculated values of the nucleation rate. At such a low relative humidity, natural ionizing radiation may play a significant role in nucleation of sulfuric acid embryos (Vohra et al. 1972). Problems mentioned by Takahashi et al. (1975) concerning aerosol modeling include the proper assumption for the particle size distribution change with time, and the effect of wall deposition and air flow pattern in future model applications to chamber studies.

Davis (1976) proposes the possible key reactions of the SO_2 conversion to sulfate via OH radicals (see Table 8) to explain the ozone bulge noted in the study of the Morgantown Power Plant (see Section V).

TABLE 8. KEY REACTIONS PROPOSED FOR THE SO_2 CONVERSION TO SULFATE VIA OH RADICALS *

(a) $SO_2 + OH + M$	→	$HSO_3 + M$
(b) $HSO_3 + O_2 + M$	→	$HSO_5 + M$
(c) $HSO_3 + HO_2$	→	$H_2SO_3 + O_2$
(d) $HSO_5 + NO$	→	$HSO_4 + NO_2$
(e) $HSO_5 + HO_2$	→	H_2SO_5** $+ O_2$
(f) $HSO_4 + HO_2$	→	$H_2SO_4 + O_2$
(g) $HSO_4 + HSO_4$	→	$H_2S_2O_8$**
(h) $HSO_4 + O_2 + M$	→	$HSO_6 + M$
(i) $HSO_6 + NO$	→	$HSO_5 + NO_2$
(j) $HSO_6 + SO_2$	→	$HSO_5 + SO_3$

* It should be noted that the detailed kinetics of only reaction (a) have been well-studied in the laboratory. Chamber studies, however, indicate that some combination of processes (b)-(j) are quite fast compared to (a).

** Both H_2SO_5 and $H_2S_2O_8$ are known to hydrolyze in the presence of water to form H_2SO_4.

The HSO_6, proposed to be produced when O_2 is added to HSO_4, oxidizes two NO molecules to reform HSO_4 which then repeats the process. This NO oxidation is postulated to explain the ozone bulge (Davis et al. 1974), but has been disputed in the literature.

Systems Applications, Inc. (Liu 1975) has developed a chemical kinetic model to study the basic features of a reactive plume from a large point source. The Reactive Plume Model (RPM) handles the transport and diffusion processes in a simple fashion, while the chemical kinetics are given more detailed attention. The considerations which were taken into account to arrive at such a model format included the exceedingly complex photochemistry prevalent in plumes where NO_x/HC ratios are high and the difficulty of solving nonlinear transport equations as compared to the ordinary controlling differential equations which describe the chemical kinetics. The following is the derived modeling equation for a plume along a wind trajectory:

$$\frac{dC_i}{dt} = \left(\frac{dC}{dt}\right) - \frac{1}{w}\left(\frac{dw}{dx}\right) UC_i - \frac{1}{h}\left(\frac{dh}{dx}\right) UC_i$$

where the rate of production of species i through chemical reactions is represented by the first term on the right-hand side, and the rate of dilution of pollutant species i within the plume along the trajectory path (x) due to horizontal spreading (w) and the variation in mixing depth (h), respectively are represented by the second and third terms on the right-hand side. A simple mass balance for the pollutant species characterizes the model equation.

A modified version of the Hecht-Seinfeld-Dodge kinetic reaction mechanism (1974) for photochemical smog formation, employing five reaction steps involving SO_2, describes the rate of change of species i due to chemical reactions. These SO_2 reactions include:

$HO_2\cdot + SO_2 \xrightarrow{k} OH\cdot + SO_3$ where $k = 1.3\ ppm^{-1}\ min^{-1}$

$SO_2 + NO_3 \xrightarrow{k} NO_2 + SO_3$ where $k = 10.0\ ppm^{-1}\ min^{-1}$

$RO_2 + SO_2 \xrightarrow{k} RO\cdot + SO_3$ where $k = 1.5\ ppm^{-1}\ min^{-1}$

$RCO_3 + SO_2 + (O_2) \xrightarrow{k} RO_2\cdot + SO_3 + CO_2$ where $k = 1.5\ ppm^{-1}\ min^{-1}$

$OH\cdot + SO_2 \xrightarrow{k} [HSO_3\cdot] \xrightarrow{O_2} HO_2\cdot + SO_3$ where $k = 900\ ppm^{-1}\ min^{-1}$

Uncertainties about atmospheric SO_2 chemistry dictate that these reaction steps remain provisional until the appropriateness of the SO_2 mechanism is better asserted by results of planned field studies.

The reactive plume model is found to be especially sensitive to variations in four of the input variables, specifically 1) plume geometry as a function of downwind distance; 2) pollutant emission rates; 3) ultraviolet radiation flux and; 4) the initial concentrations of NO, NO_2, SO_2, O_3, and hydrocarbons.

Huang et al. (1974) followed the methodology of Hazbun et al. (1971) in the Tennessee Valley Authority's derivation of a kinetic model designed to provide a detailed account of the rates of change in concentrations of the various components resulting from their chemical interactions and the effect of meteorological conditions upon these reactions. The Hazbun approach was to construct profiles by measuring the concentrations of the various species under differing meteorological conditions at a number of downwind distances. On the basis of then available literature, a kinetic model was devised for probable chemical reactions and various rate constants determined from experimental data were selected to formulate a set of rate equations. The set of rate equations was integrated to give the theoretical profiles of the various components. The chemical reactions possible (from a literature search at that time) were upwards of 60; however, only 29 reactions were considered important from considerations of the magnitudes of the rate constants and reactant concentrations. Unfortunately, exclusion of the OH + SO_2 reaction and numerous updated values of the rate constants have basically antiquated this model.

Model validation studies have not as yet been conducted in the atmosphere since data in sufficient detail to validate model predictions in plumes are simply nonexistent.

A numerical model has been developed for predicting precipitation scavenging of reactive pollutants from power plant plumes (Dana et al. 1976). The Scavenging Model Incorporating Chemical Kinetics (SMICK) can calculate collection, liquid-phase chemical reaction, and (possible) desorption of multiple plume-bound pollutants as they interact with falling raindrops to be ultimately deposited on the ground. A schematic of these physical interactions is shown in Figure 4. SMICK also possesses the capability to perform calculations for any specific aqueous-phase kinetics mechanism expressed in a proper subroutine form. Initial applications of the Scott and Hobbs (1967) aqueous-phase oxidation mechanism for SO_2 (which depends strongly on the presence of dissolved ammonia) showed that in comparisons of these predicted results with field measurements, taken at the Centralia power plant (see Section V), the mechanism is not adequate to account for the sulfate formation rates observed. To calculate washout concentrations, the following equation is utilized:

$$\frac{dc}{dz} = \frac{3K_y}{aV_z} (Y - H'c)$$

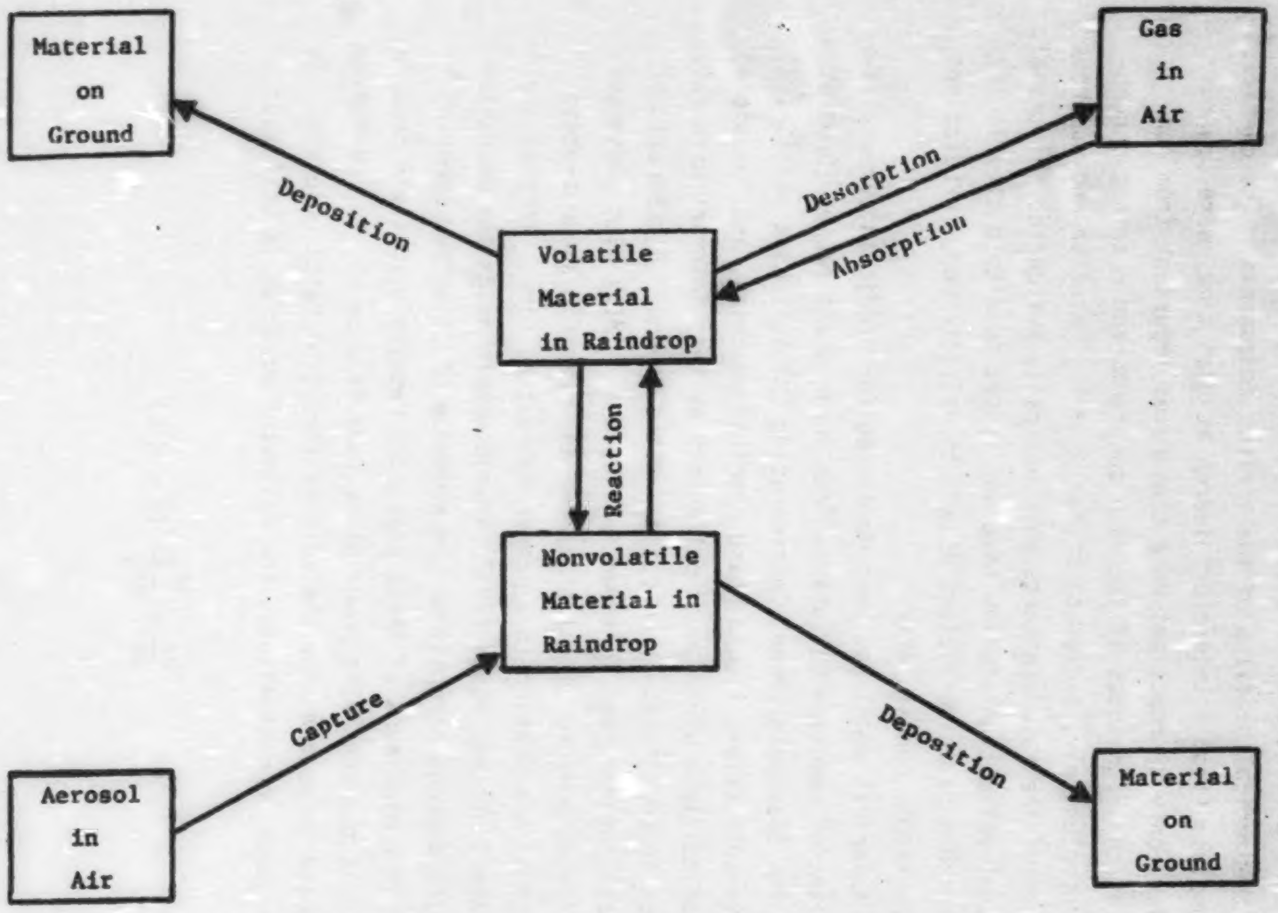
Material
on
Ground
Gas
in
Air
Deposition
Desorption
Absorption
Volatile
Material
in Raindrop
Reaction
Nonvolatile
Material in
Raindrop
Capture
Deposition
Aerosol
in
Air
Material
on
Ground

Figure 4. Schematic of Physical Interactions Modeled by SMICK (from Dana et al. 1976).

where

c = the liquid phase concentration (moles/cm^3)
z = distance above ground at stack base (cm)
K_y = overall mass-transfer coefficient (mole/cm^2-s)
V_z = the terminal fall velocity of a raindrop of radius a (cm/s)
a = radius of a raindrop (cm)
Y = mole fraction of pollutant a in local gas phase
H' = solubility parameter (cm^3/mole).

Provided values of K_y, V_z, Y, and H', the above equation is numerically solved for a number of selected drop sizes; the results are then distributed according to the raindrop spectrum to obtain average concentrations. In addition to performing this function, the computer code allows for a number of sophistications including nonlinear solubility behavior, lofting plumes and nonvertical rainfall.

A steady-state, one-dimensional mass transfer model describing the dry deposition process has been developed by Sehmel et al. (1973) and has been employed by Sehmel and Hodgson (1974) to predict particle deposition velocities to simple surfaces. The minimum experimental data required to successfully employ the model are friction velocity, roughness height, particle size distributions, and atmospheric stability. Conceptually, the model is a three-box simulation of particle transport utilizing the continuity equation. The three elements consist of:

Box 1 - The atmospheric turbulent layer in which the transfer processes are best described by micrometeorological eddy diffusivity.

Box 2 - A layer just above the vegetative canopy or surface elements in which the transfer processes are modified by the presence or structure of the canopy or surface.

Box 3 - A layer (occupied by the canopy or surface elements) in which the final transfer process is best expressed by surface mass transfer coefficients, where the interaction between the surface material and the pollutant is important.

The basic model assumptions, as stated by Sehmel and Hodgson (1974) are:

1. Particles diffuse at a constant flux from a uniform concentration of particles.
2. A relationship for particle eddy diffusivity can be determined.
3. The effect of gravity can be described by the terminal settling velocity.
4. Particle agglomeration does not occur.
5. The particles are completely retained by the surface.

With the basic model assumptions, the deposition flux to a surface is described by

$$N = -(\varepsilon + D)\frac{dC}{dz} - V_t C$$

where

N = deposition flux to environmental surface (particles $cm^{-2}s^{-1}$)

ε = particle eddy diffusivity (cm^2s^{-1})

D = Brownian diffusivity (cm^2s^{-1})

C = particle concentration (particles cm^{-3})

z = height (cm)

V_t = absolute value of terminal settling velocity (cm s^{-1})

This equation is then integrated to provide integral diffusional resistances, which may be related to deposition velocity. Predicted deposition velocities were graphically presented for a 1 meter reference concentration height, for stable and unstable atmospheres, with the parameters for each figure being either friction velocity (range of 10 to 200 cm s^{-1}) or roughness height (10^{-3} to 10 cm). In general, deposition velocities increase with an increase in either friction velocity or roughness height parameters. For all combinations of these two parameters the minimum deposition velocities were found to occur in the particle diameter range of 0.04 to 0.4μm. Deposition velocities were determined to be nearly independent of particle diameter only in the 0.1 to 1.0μm size range (note; this is the "accumulation mode" where 80 percent of atmospheric sulfate is found (E.P.A. 1974)).

Horst (1974) developed a surface depletion model which improves on the usual Gaussian plume model "source depletion" representation of loss of airborne contaminant due to deposition. The Gaussian source depletion approach appropriately reduces the source strength as a function of downwind distance. In the Horst approach, the air concentration at any location can be calculated as the sum of the nondepositing diffusion from a primary source at coordinates (0,0,h) plus the diffusion from all of the upwind surface sources which account for deposition. The derived equation for the source depletion model considering the crosswind-integrated air concentration is:

$$\overline{\chi}\ (x,z) = \overline{D}\ (x,z,h)\ \left(Q_o - \int_o^x V_d\ \overline{\chi}\ (\xi,\ z_d)\ d\ \xi\right)$$

where

$\overline{\chi}$ (x,z) is the crosswind integrated air concentration

Q_o is the undepleted source strength at x = o

V_d is the deposition velocity (constant of proportionality)

ξ represents deposition coordinates at downwind point (x,y,z)

Z_d is the reference height

$\overline{D}$ is a diffusion function.

The Horst model and its predecessor were compared for a variety of conditions to judge the efficiency of the source depletion model with respect to the surface depletion model. Calculations were made for a variety of source heights (2 m, 10 m, and 100 m) and for three Pasquill stability categories (A, D, and F). It was determined that the differences between the two models increase with increasing stability. The source depletion model was characterized by an overprediction of total deposition and a consequent underprediction of the remaining airborne material. In one case of moderately strong deposition and stable conditions, the source depletion model for parameters at a downwind distance of 10 km was found to be in error by factors of 3 and 4. At greater downwind distances and in cases of stronger deposition, these factors can increase substantially. The source depletion model was found to be entirely sufficient for low deposition conditions but to increase in error for high deposition case. For example, for $V_d/\overline{u} = 10^{-3}$ the source depletion model is in error by 10 to 20 percent for three release heights and three stabilities; for $V_d/\overline{u} = 10^{-2}$ and unstable thermal stability the greatest error is approximately 35 percent.

Droppo (1974) described the types of input necessary for developmental modeling of dry deposition processes on high and low vegetation canopies. Four regimes characterize the model of overall deposition over a canopy, these include 1) air layer above the canopy; 2) air-vegetation layer within the canopy; 3) the surface layers over the individual plant elements and 4) inside the plant elements where internal processes limit the rate of assimilation of certain materials into the plant. With the assumption that atmospheric and canopy characteristics can be expressed as height and time functions, the functional relationship for a given site is:

$$r(z,t) = r_1(z,t) + r_2(z,t) + r_3(z,t)$$

where

r is the total resistance (to flow of material)

r_1 is the resistance in the first regime

r_2 is the resistance in the second regime

r_3 is the resistance in the third regime

There is no defined r_4, since these processes will directly affect the third regime.

The model input variables consist of a series of plant characteristics and processes (functions of height and time). These are: Physical Characteristics, such as canopy height, density of foliage, canopy roughness and leaf area index; Physiological Characteristics, such as stomata openings and surface characteristics and finally Concurrent Fluxes such as momentum, latent heat, and sensible heat. No quantitative data were provided in the Droppo (1974) paper, but a series of qualitative results were given. The deposition process is expected to differ depending on physical characteristics of the canopy; greater amounts of deposition will occur in the lower regions of the low canopies than in higher less dense canopies. The upper portions of higher vegetation canopies are the locations for significant deposition of material. Results of diffusion, wind profiles, energy budget and physiological research relating to vegetation canopies will be incorporated into this model as it is developed.

Section V

FIELD STUDIES OF PLUMES

Data have been gathered on plume transformation chemistry, and theories have been developed and tested, in field studies of specific plumes. The methods and results of a number of these studies are discussed below.

COLBERT POWER PLANT

In pioneering work of the use of the helicopter as a plume tracking tool, a limited field study program was undertaken at the TVA's coal burning Colbert Power Plant to measure the rate of atmospheric oxidation of SO_2 during different weather conditions (Gartrell et al. 1963). At that time the plant, located approximately 20 km southwest of Wilson Dam, Alabama, had four 200 mw units, each with a 97 m (300 feet) stack. About 70 percent of the flyash was removed by mechanical flyash collectors. The reported results of earlier laboratory studies were utilized in the planning of this project to provide the investigators with a basis of comparison and indications of what to expect from the field study.

Sampling from a helicopter was designed to directly determine SO_2 and SO_3 components, utilizing a filter paper separation technique. (The instrumentation at that stage of the technology was not adequate for making SO_3 or H_2SO_4 measurements; unfortunately reliable measurements of ambient H_2SO_4 levels are still not a reality even today.) Auxiliary instrumentation included the altimeter, a spring-wound clock, and in later tests, wet and dry temperature probes. A voice recorder was utilized to store data from these instruments for subsequent analysis. Measurements were made during early morning inversion conditions for easy plume detection and presence of maximum SO_2 plume concentrations. Close to the plant (1 to 2 miles), longitudinal flights were made along the centerline of the relatively narrow plume to shorten the total sampling time necessary for each sample. Further away from the plant, where the plume was wider, the helicopter conducted replicate flights across the centerline of the plume in progressive sections. A data summary of the flights (Table 9), taken from the Gartrell et al. (1963) study, depicts the range of SO_2 oxidation rates encountered for a variety of different time, distance, and relative humidity conditions. The oxidation rates are seen to range from 0 to 3.7 percent for measurements characterized by low humidity, to rates of 21 and 32 percent for humidities close to 75 percent. An SO_2 oxidation rate of 55.5 percent for a time of 108 minutes was observed in one sample run. A common starting time for calculating oxidation rates was not a condition of the sampling methodology.

TABLE 9. SO_2 OXIDATION STUDIES - COLBERT STEAM PLANT PLUME (from Gartrell et al. 1963)

Date 1960	Sample No.	Travel from Point of Emission		Relative Humidity in Plume (Percent)	SO_2 Oxidation (Percent)
		Time (min)	Distance (miles)		
		"Low Rates"			
8/2	1	5	.25-1		0
	2	5	.25-1		0
	3	5	.25-1		1.20
	4	15	1-1.5		0
9/2	1	30	2-3		3.70
	2	78	8		2.20
10/14	1	12	.5-1.5	62	2.15
	2	60	5-6	54	3.23
10/26	1	6	.25-1.25	45	1.50
	2	84	8-9	48	2.70
10/28	1	12	.5-1.5	68	1.10
	2	84	8-9	70	4.10
		"High Rates"			
5/3	1	13	1.1		13.80
	2	13	1.1		10.00
	3	13	1.1		19.20
8/19	1	108	8-10		55.50
	2	23	.75-2		8.00
10/11	1	12	.5-1.5	74	21.60
	2	96	8	73	32.00

Gartrell et al. (1963) state that the limited data obtained in their study do not provide a basis for accurate estimates of the post-emission absolute rate of SO_2 oxidation. The investigators postulate that the dominating factor controlling the oxidation rates was moisture within the plume or ambient strata; especially at relative humidity values greater than 70 percent. Newman et al. (1975) contend that upon closer inspection of the Gartrell et al. (1963) data, a dependence on humidity is not that clear, and attribute the observed high SO_2 oxidation rates to very high particulate loading in the plume.

The TVA's Colbert plant was selected once again for the collection of data on the characteristics of a coal-fired power plant plume, including the chemical constituents and their concentrations with respect to downwind distance (Huang et al. 1974). A kinetic model of plume chemical interactions was developed from data gathered, and has been described in Section IV. At the time of the field program late in 1972, the plant was characterized by a 980 mw capacity, with three stacks, two at 93 m (300 feet) and one at 152 m (500 feet).

A Sikorsky model S-58 helicopter was equipped with instruments designed to measure sulfur dioxide, total sulfur, nitrogen dioxide, nitric oxide, ozone, nitrous acid, toluene, particles, humidity and temperature for the project duration. During the program, the helicopter would leave the TVA airport in Muscle Shoals, Alabama, at sunrise and fly to the vicinity of the Colbert plant. During the flight to the steam plant the background (ambient) concentrations of the pollutants were measured. Upon arrival at the plant a vertical temperature profile was made to an altitude of 1200 m (4000 feet). Once the temperature profile was completed, plume measurements were initiated across the plume centerline at predetermined distances downwind. Samples were taken and continuous gas analysis was started when the helicopter entered the plume. A minimum of 60 seconds in the plume was allowed for plume sampling; circling within the plume was required for samples in the narrow portions of the plume. To allow the sampling probes to function properly, an airspeed of near 35 knots was maintained during a sampling run. The sulfur analyzer was continuously operated and utilized as an indication of plume boundary location. Temperature and dew point were measured simultaneously with the pollutants, and before entering the plume. The plume traverses were continued until the morning inversion was completely broken and the plume was too dispersed to measure. A second vertical temperature profile was made at the end of the sampling periods. The traverses of the plume by the helicopter were made at downwind distances of 0.8, 1.6, 3.2, 6.4, and 9.5 kilometers. The plume was traversed at distances greater than 16.1 km (10 miles) when the atmosphere was extremely stable. Measurements of ground level meteorological parameters, specifically dry and wet-bulb temperatures, solar radiation, and wind speed and direction were made concurrent with the airborne studies. Winds aloft were determined by releasing and tracking pibals.

The gathered experimental data were utilized to construct concentration profiles. Theoretical concentration profiles were constructed by the integration of 35 rate equations for the kinetic model chemical reactions assumed to be the most likely to occur. These theoretical profiles were altered by varying the rate constants until the resulting profiles agree as closely as desired with the observed profiles, and these were taken to be a likely description of the plume chemistry. However, it appears that the 35 reactions used by Huang et al. form a weak representation of reality. The reactions of radical chemistry were poorly represented; rate constants could be estimated with an accuracy of no more than an order of magnitude.

Due to the suspected inaccuracy of the experimental results, the overall rate of oxidation of SO_2 could not be determined from either (1) the change in concentration of SO_2 by separating the effects of dispersion and oxidation or (2) a comparison of concentrations of SO_2 and sulfates. SO_2 oxidation rates appeared to be slow in any event. The authors expressed the desire that the results in their report should not be accepted as quantitative due to the limitations involved.

CRYSTAL RIVER POWER PLANT

A series of investigations at the Florida Power Corporation's coal burning Crystal River Power Plant, utilizing aircraft, had a goal of testing a method for evaluating SO_2 decay rates in the plume (Stephens and McCaldin 1971). The 375 MW capacity plant has a single 152 m (500 ft.) discharge stack and is situated on the flat Gulf Coast plain 129 km (80 miles) north of Tampa, Florida.

To differentiate between a decrease in SO_2 concentration due to atmospheric dispersion processes and a decrease in concentration due to oxidation or any other chemical or physical decay factors, a technique believed by Stephens and McCaldin (1971) to be "conservative" in nature was utilized. The technique consisted of a comparative ratio of sulfur dioxide concentration to a conservative tracer concentration (consisting of submicron-sized particulate matter emitted from the stack); supposedly the ratio of particulates to SO_2 would approximate a constant as the plume aged and diffused, if the SO_2 were conservative. The ratio would increase as the plume ages if the SO_2 demonstrates a measurable decay rate. Such a procedure has been challenged by Friend et al. (1972), who explained that particulates cannot be used as a conservative tracer since the sulfate particles formed as a result of the oxidation of sulfur dioxide can significantly contribute to the total mass.

It was determined that the time required for the decay of one-half of the sulfur dioxide indicated a first order rate equation for the reaction. The range of SO_2 loss rates were described to vary from a negligible value at low relative humidity (30-40 percent), to a half-life of approximately 140 minutes at medium relative humidity (40-50 percent), and 70 minutes at high relative humidity (78-80 percent).

MORGANTOWN POWER PLANT

Field studies at the Morgantown Power Plant of the Potomac Electric Power Company were conducted by Davis et al. (1974) to determine the extent to which NO_x and SO_2 chemistry occurs as a function of distance from the emission source. The plant, which burned an oil-coal fuel mixture (75 percent residual #6 oil and 25 percent coal) is located 64 km (40 miles) south of Washington, D.C., and has a 1250 MW capacity. The collection of field data, which took place from October 1973 to August 1974, was accomplished through use of a single engine Cessna 205 airplane early in the program and a Cessna 172 later in the program. Extensive field data were collected from instrumentation aboard the Cessna 205 aircraft which included a Monitor Labs 8410 chemiluminescence ozone analyzer, a Monitor Labs 8440 chemiluminescence $NO\text{-}NO_2\text{-}NO_x$ analyzer and a Meloy SA-160R flame photometric SO_2 analyzer. Instruments designed to monitor CO and hydrocarbons were also employed. Horizontal cross-sectional traverses were made of the plume at altitudes ranging from 213 to 914 meters at various downwind distances up to 56 km. A major observation in the field program was the depletion of ambient ozone within the plume near the stack at distances of 5 to 8 km, and an increase in ozone at 50 km downwind with a decrease in plume SO_2. The plume depletion features close to the plant are illustrated in Figures 5a and 5b from the Davis et al. (1974) report. An increase of roughly 30 percent over ambient ozone levels was noted at 56 km, but the profiles at this downwind distance were not available. The details of the atmospheric chemistry theory proposed by Davis et al. (1974) to explain the net generation of ozone in plumes have been described earlier in Section IV and will not be repeated here.

KEYSTONE AND NORTHPORT PLANT

The extent of oxidation of sulfur dioxide to sulfate was examined in a study of the Pennsylvania Electric Company's Keystone coal-fired power plant by Newman et al. (1975a), and the Long Island Lighting Company's Northport, New York oil-fired power plant by Newman et al. (1975b). The Keystone Plant generates a total of 1800 MW using two boilers whose units are equipped with electrostatic precipitators rated at 99.5 percent efficiency,

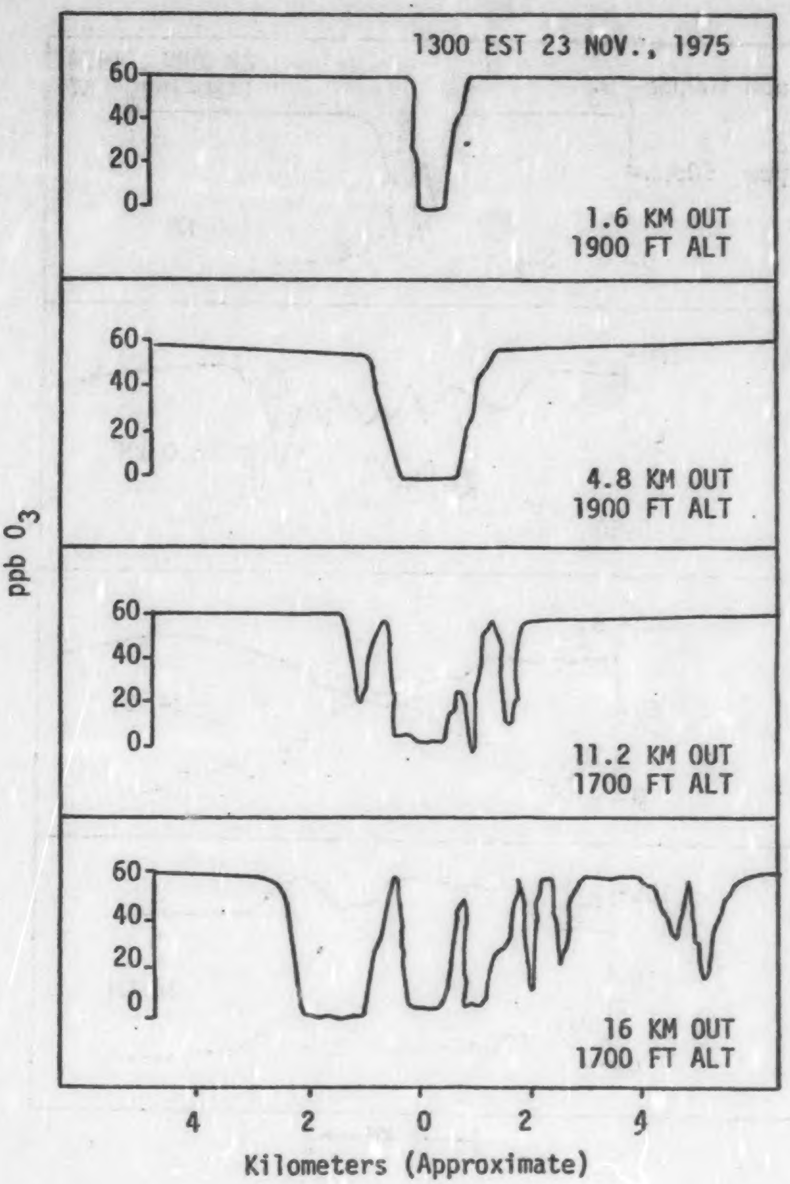

Figure 5a. Profiles of Crossplume Ozone Concentration at Various Elevations Downwind of the Morgantown Power Plant (from Davis et al. 1974a).

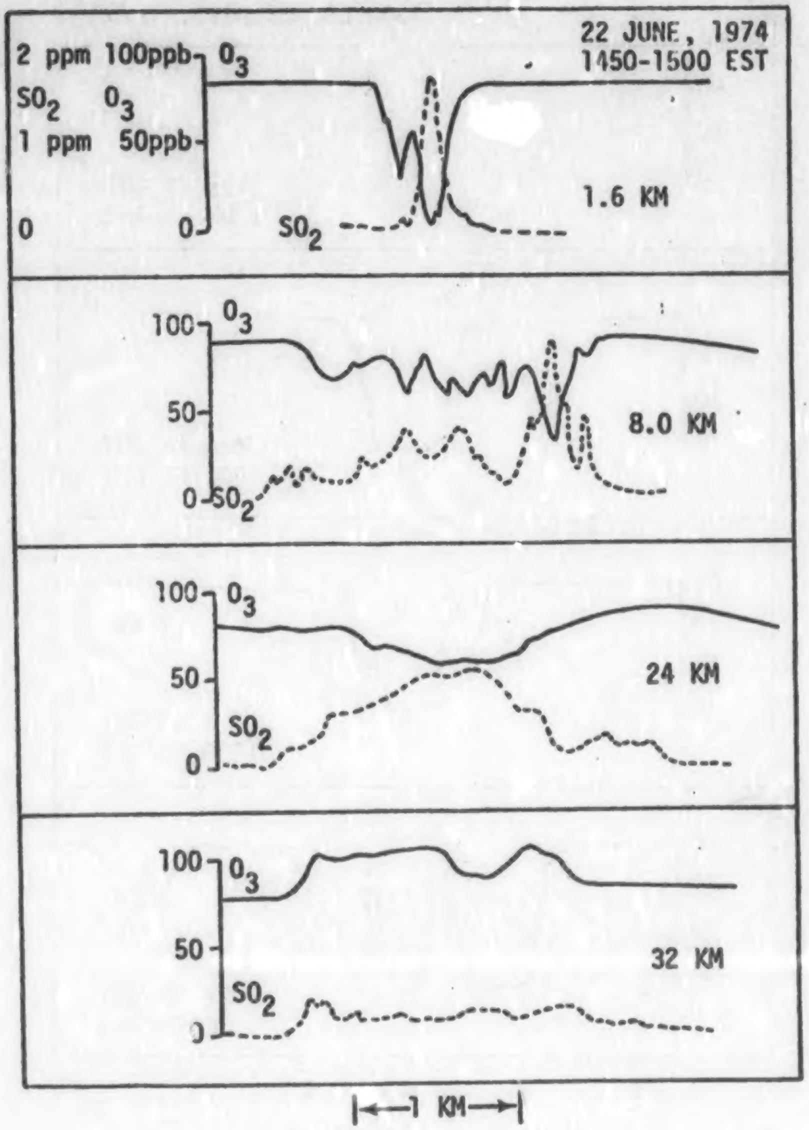

Figure 5b. Profiles of Sulfur Dioxide and Ozone Crossplume Concentrations at 600 m Altitude at Various Downwind Distances from the Morgantown Power Plant (from Davis et al. 1974).

each unit served by a 244 m stack. At Northport, the 760 mw plant operates with two boilers, each with a 183 m stack. Mechanical cyclone type precipitators on these units have a rated efficiency of 85 percent.

In both plant studies, a single-engine Cessna 182 was utilized to obtain plume samples for isotopic ratio and concentration measurements of the ^{32}S:^{34}S naturally occurring in the fuel sulfur. The aircraft was instrumented with a hi-vol for sampling of sulfate (on fiberglass filters) and SO_2 (on alkaline impregnated filters), an electroconductivity SO_2 analyzer for locating the plume, and auxiliary equipment for measuring temperature, relative humidity, altitude, airspeed and aircraft location. Background measurements of SO_2 and sulfates were obtained upwind of the plume at plume altitude and repeated crosswind traverses at a minimum of five distances (when feasible) were made in the plume during data gathering. Sulfate sampling under the plume was conducted at two distances downwind.

Concentrations of the conservative tracer SF_6, compared to SO_2 as a function of distance, were used to assess the SO_2 losses by oxidation and deposition. Hi-vol samplings of sulfate particulates were so small that a reliable analysis could not be conducted to fit the data obtained to the oxidation process postulated for both coal and oil-fired plumes; measurements of SO_2 converted to sulfate were therefore based upon the assumption that reductions of SO_2 concentrations greater than those found for SF_6 should be attributed to conversion to $SO_4^=$.

The fraction of SO_2 converted at measurement points in the plume downwind from the stack was also calculated from the expression:

$$(1 - f) = \frac{\delta SO_2\ (\text{fuel}) - \delta SO_2}{1000\ \alpha}$$

where

f = the fraction of SO_2 converted

δSO_2 = the "del value" which is a measure of deviations of the ratio $^{32}S/^{34}S$ at the point of measurement from a standard isotopic sulfur ratio for $^{32}S/^{34}S$ of 22.210

α = k-1, where k is the equilibrium constant between SO_2 and SO_3 for the reaction

$$^{34}SO_2 + {}^{32}SO_3 \overset{k}{\rightleftarrows} {}^{32}SO_2 + {}^{34}SO_3.$$

Eleven standard plume sampling runs were made at the Keystone power plant. The SO_2 concentrations were measured in the flue gas at the stack and at six downwind distances. The data obtained on these runs were combined, normalized and averaged with the results summarized in Table 10.

TABLE 10. NORMALIZED AND AVERAGED DATA FROM 11 PLUME SAMPLING RUNS MADE AT THE KEYSTONE POWER PLANT (from Newman et al. 1975a)

Downwind Distance from Plant (km)	0.0	0.8	1.6	3.2	4.8	16.1	48.3
δ * (del value)	2.8	2.5	2.1	1.9	2.2	2.2	2.1
Percent SO_2 conversion to $SO_4^=$ based on δ calculations	0.0	1.3	3.6	4.4	3.0	3.0	3.5
Percent SO_2 conversion to $SO_4^=$ based on measured concentrations	0.0	3.0	4.1	3.6	1.8	0.3	0.0

* The del values are normalized to the average flue gas value of 2.8.

The percents of SO_2 conversion shown in this table refer to conversion in the atmosphere after leaving the stack, and to a common base of 0.0 percent at the stack. Reductions in conversion with increasing distance were attributed to removal of $SO_4^=$ from the atmosphere after its formation.

The following observations were obtained from these data:

1. The indicated decrease with distance in percent SO_2 converted based on concentration supports evidence for the apparent dropping out of sulfate from the plume.
2. The percent SO_2 conversion to sulfate based on isotopic ratio measurement tends to attain a steady value of 3 to 4 percent, which is independent of time or distance; however, the authors point out that super-imposed on

> these percentages, which are depicted as a function of distance, are wind velocities that ranged over a factor of four. Part of the variability may be associated with a large range in the isotopic ratio of the sulfur in the coal at the plant.

The low SO_2 conversion rates of less than 5 percent are attributed to a rate limiting process. This is supported by the low particulate content of the Keystone plant plume, on the order of 10^5 μg/m^3.

A calculated SO_2 plume oxidation half-life of 10 hours was derived from a humidity-independent second order kinetic expression, and from a resulting rate constant of 1 ppm^{-1} hr^{-1}. The oil-fired plume had no reported particulate loadings. Vanadium particulates, originating from fuel oil were postulated (by Newman et al. 1975b) to be the catalyst in a mechanism whereby the SO_2 is in equilibrium with water on these particulates and catalytic oxidation subsequently occurs. Newman et al. (1975a) did not postulate a similar mechanism involving Fe_2O_3 in the coal-fired particulate matter. The noted highly variable SO_2 conversion in both oil- and coal-fired plumes and the lack of a definite correlation of the percent SO_2 conversion with relative humidity are major research outcomes of these studies.

FOUR CORNERS PLANT

Further evidence for low SO_2 oxidation rates in coal-fired power plant plumes was provided by the University of Utah Research Institute (1974) in a study carried out at the Four Corners power plant in New Mexico. The program objectives were to measure the degree of SO_2 conversion and to evaluate its impact on concentrations of atmospheric sulfate and visibility. Parameters measured to determine the conversions of SO_2 to particulate sulfate included particulate ammonium sulfate; upwind and downwind difference in concentrations compared to ambient SO_2, and the speed of wind. One test resulted in a 0.37 percent hr^{-1} conversion of SO_2 to particulate sulfate at about 21 km downwind, and relative humidities of 30-51 percent. Another test resulted in a 0.76 percent hr^{-1} conversion 21 km downwind with relative humidities of 26-40 percent. Conversions of 0.45 percent hr^{-1} at about 60 km downwind were noted in this same test. Both tests were characterized by SO_2 concentrations ranging from 0.01-0.06 ppm. This study possibly provides a more realistic measure of plume SO_2 oxidation rates due to the fact that the SO_2 rates of oxidation provided are the result of direct, positive sulfate analysis and not deduced from the comparison of SO_2 loss with plume tracer material.

MISTT PROGRAM

Extensive studies involving three-dimensional mapping of large plumes from multiple urban sources were accomplished in the St. Louis area as part of the Regional Air Pollution Study (RAPS) and the Midwest Interstate Sulfur Transport and Transformation Study (MISTT) during the summers of 1973-1975 and during February of 1975 (Wilson et al. 1976). These studies challenged the adequacy of the usual technique of calculating the fractional conversion of SO_2 to sulfate from SO_2 concentration and sulfate data obtained by aircraft flying through the plume on a variety of flight paths. Based on observational data of typical cross-plume profiles of Aitken nuclei concentration, light scattering aerosol, SO_2 and ozone, it is believed that the SO_2 conversion rate is different in various parts of the plume, the higher rates favoring the plume edges rather than center, although this difference is not evident in all cases. The conclusion reached by Wilson et al. (1976) is that measurements of SO_2 and sulfate mass flow rates in the plume are the best indicators of conversion rate.

Aircraft mapping of the plumes of the 2400 MW Labadie plant, emanating from three stacks 213.5 m (700 ft) tall located west-southwest of St. Louis was conducted out to plume distances approaching 100 km. The basic concept was the measurement of the gaseous and particulate sulfur flow rates at increasing distances from the source to derive pollutant budgets and ultimately to allow estimates of the rate at which transformations and removal processes act on the individual pollutants. The coordinated efforts of two instrumented aircraft, an instrumented van and three mobile pilot balloon units were utilized in the plume mapping program.

Plume aerosol volume flows were calculated from aircraft size distribution measurements with an electrical aerosol analyzer and an optical particle counter made during horizontal passes and from upwind spirals through the plume. The results of plume aerosol flow rates and calculated sulfur conversion obtained for one test day showed little sulfate formation in the first 30 km, and a conversion of some 2% hr^{-1} in the region 30-50 km from the source. An important observation of the MISTT studies is low $SO_4^=$ formation until NO is converted to NO_2 and then subsequent higher $SO_4^=$ formation: this stresses the necessity of long distance measurements from the stack source, since at closer distances (i.e., 16-32 km) only the stack sulfate, derived from SO_3 from in the combustion process, may be present.

SO_2 DEPOSITION STUDIES

Pacific Northwest Laboratories (Nickola and Clark 1974) designed a technique for field measurements of deposition of a particulate plume by tracking with an inert gas plume. The inert gas, Krypton-85, was simultaneously released with fluorescent zinc sulfide particulate tracer from a single site 26 m in elevation; the air concentrations of both plumes were examined downwind of the release point. The mass median diameter of the zinc sulfide was 5 μm; removal processes such as impaction and gravitational settling became important if the particle diameter was greater than this value (Dabberdt and Smith 1975). The deposition process can be credited with the expected deficit of mass in the particulate plume as both chemically stable plumes travel downwind; an estimation of the amount of zinc sulfide deposited was conducted by a graphical study of the differences between the two vertical profiles of concentration near the ground. The field sampling grid was comprised of more than 600 filters at 1.5 m height placed about the tracer release tower in a concentric fashion, on sectors of six arcs at distances of approximately 0.2, 0.4, 0.8, 1.2, and 3.2 km* from the point of release. The results showed that under moderately stable conditions about 7 percent of the particulate plume was deposited during passage to a distance of 842 m. Zero to 1 percent of the particulate tracer was deposited within 812 m from the release point under more stable atmospheric conditions. Approximately 1 percent of the zinc sulfide tracer is estimated to have touched down within 200 m of the source during a case characterized as flat with sagebrush and steppe grasses dominating the region.

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* Arcs at 0.8, 1.6 and 3.2 km were actually concentric about a point 100 m due south of the release point. Thus, the distance from release point to arc varies with azimuth.

Garland (1974) has measured the rate of SO_2 dry deposition both in the field and in the laboratory using $^{35}SO_2$, a radioactive tracer as one technique and the concentration gradient method as the other. Several of the field efforts, utilizing the radioactive tracer method to measure the deposition velocity to grass, consisted of releasing a small amount of SO_2 containing 50 millicuries ^{35}S over a 30-minute period at a height of 1 m, 50 m upwind of an arranged sampling area near the center of a large field. $^{35}SO_2$ concentrations were determined by air bubbler samplers mounted above marked plots. Exposure samples of the grass were removed from several of these plots and the uptake of ^{35}S was resolved by transferring the total extracted sulfate to a liquid scintillation counter. The results were interpreted as a deposition velocity defined by:

$$Vg = \frac{\text{radioactive particle counts in grass per unit ground area}}{\text{concentration in air X time of exposure}}$$

The gradient method, when used to determine the flux of SO_2, assumes that momentum, heat, and SO_2 fluxes are constant with height over the range of measurement. An area of uniform surface extending for at least 100 times the measurement height upwind is required. The diffusivity of momentum or the diffusivity of heat are used as approximations of the flux of SO_2; the diffusivity for momentum was used in the Garland (1974) study. These diffusivities are defined as:

$$Km = \frac{ku^*(z-d)}{\phi_M} \text{ or } K_H = \frac{ku^*(z-d)}{\phi_H}$$

where

Km = diffusivity of momentum (m^2 s^{-1})
K_H = diffusivity of heat (cal m^{-1} s^{-1})
k = von Karman's constant, an empirical constant of value 0.4
u* = friction velocity defined in terms of shear stress on the surface due to the wind (m^{-1})
z = height above surface (m)
d = reflects the effect of tall crops in elevating the height of the momentum sink above the surface (m)
ϕ_M = the dimensionless shear of momentum
ϕ_H = the dimensionless shear of heat.

The results of three sets of field measurements of the dry deposition of SO_2 over grass vary depending on the time of year and characteristics of the canopy. For short grass in March and June, the mean Vg was 0.55 cm s^{-1}; medium grass (radioactive method) in June-November gave a mean Vg of 1.19 cm s^{-1}; using the gradient method from November - January over medium grass gave a mean Vg of 0.77 cm s^{-1}. Over long grass, using the radioactive method, a mean result of 0.85 cm s^{-1} was found; this compares to a Vg of 0.61 cm s^{-1} for 14 measurements reported by Shepherd (1974), and to the deposition velocity of 0.4 cm s^{-1} over a wheat field found by Fowler and Unsworth (1974).

Most of the research concerned with the absorption of atmospheric water soluble gases by vegetation has been conducted using environmental chambers constructed to simulate different field environment conditions. In the research of Hill and Chamberlain (1974), two environmental chambers were designed that accomplished this simulation by controlling wind velocity, CO_2 concentration, temperature, relative humidity, light intensity, and pollutant concentration. The standard alfalfa canopies used in the chamber were found to result in SO_2 and NO_2 concentration profiles that indicated that these substances were efficiently removed from the atmosphere by the surface and subsurface vegetation of the upper portion of the plant canopy. The rate of pollutant removal by vegetation was shown to be affected by wind velocity, canopy height, and light intensity, with the removal rate of pollutant found to increase linearly with the increase of pollutant concentration until those pollutant levels when serious physiological affects take over the plant. Due to the lack of actual field data, the authors describe a hypothetical situation in which they attempt to evaluate the role of vegetation as a sink for air pollutants in any specific area. A large coal-fired power plant was assumed to produce average ambient SO_2 concentrations of about 6 ppb within 32 km of it. Vegetation cover in this area, with an average SO_2 uptake rate equivalent to that of a 40 cm alfalfa canopy ($17\ \mu l\ min^{-1}\ m^{-2}\ pphm^{-1}$) during daylight hours, would give an uptake rate of 1.24×10^5 kg/day in the $32.5 \times 10^8\ m^2$ within 32 km of the plant. This would be 49 percent of the SO_2 emitted from the plant; the percentage removed would be greater if the area were a forest, or less if the vegetation were scarce. The authors bring up an important point, relating the conversion of SO_2 to $SO_4^=$ in the atmosphere to vegetation pollutant sinks. The low conversion rate of SO_2 to $SO_4^=$ particles, in rural areas where photochemical smog reactions are less important, will result in high percentages of SO_2 removed by vegetation before it is oxidized to sulfate, if vegetation acts as an SO_2 sink.

PRECIPITATION SCAVENGING STUDIES

To assess the extent of depletion from a diffusing plume, an experimental investigation was conducted at the Keystone Power Plant (located in a polluted area) as part of the LAPPES program to measure the sulfur dioxide washout by natural precipitation (Hales et al. 1971). Precipitation samplers were strategically placed on a surface sampling line which extended across the overhead plume. The compound of primary interest was SO_2, although sulfate, nitrite, and nitrate levels were also determined from selected samples. Samples were delivered to a mobile laboratory for analysis as quickly as possible upon their collection. The mass of water-soluble particulate recovered from each precipitation collector indicated the degree of pollutant deposition from the plume through precipitation scavenging and dry deposition. The sulfate and nitrate results exhibited a large amount of scatter which according to the authors was suspected to have arisen due to artifacts within the samples. The sulfur dioxide distributions observed in the experiments were not sufficiently well-defined for the calculation of washout coefficients. Observed values of SO_2 washout flux were stated to be less than theoretical estimates by factors of 50 to 100. Notable was the absence of a positive correlation of washout sulfur dioxide concentration with the location of the Keystone plume, attributed to the plume shallowness at short distances from the source which permits the least time for sulfur dioxide sorption and the greatest time for its desorption. In several cases, a negative correlation of washout beneath the plume was discovered. The results indicated that the SO_2 scavenging process was more complicated than the investigators had suspected. The conclusions reached, based on the data obtained, were as follows:

1. The actual removal rate of SO_2 from the plume by the washout mechanism from the stack source at 244 m (800 ft) above ground level was up to two orders of magnitude less than predicted by preexisting theory. Washout fluxes in rainfall and snowfall were clustered about a value of 10 µmoles $(m^2 hr)^{-1}$ for precipitation rate normalized to 1 mm hr^{-1}.

2. A factor of two difference was noted from the relationship between the observed washout and precipitation rate and the relationship derived from preexisting theory. More efficient washout by large drops than is predicted by preexisting theory is attributed to the larger exponent of precipitation rate in the observed relationship.

3. Increases in washout concentrations of SO_2 were apparent with distance from the stack. Lateral diffusion of the plume, according to preexisting theory, is responsible for decreases with distance of concentrations proportional to the vertically integrated mass of sulfur dioxide.

4. Washout of background sulfur dioxide concentrations appeared to interfere strongly with plume-washout observations. Remote sources about 20 km to the southeast of the Keystone plant contributed heavily to the SO_2 washout observed.

5. There were indications that sulfur dioxide washout was inhibited in some way by the Keystone plant or that the SO_2 was converted to sulfate. A depression of SO_2 concentration observed in precipitation collected beneath the plume under certain circumstances when the washout of background sulfur dioxide was relatively high supported this.

Three experimental studies of the washout of power plant effluents were conducted at the State of Washington's Centralia coal-fired plant (Dana et al. 1975, 1976). The Centralia stacks have a height of 143 m above the ground. Heavily forested hills, rising to within 50 km of the stack tops comprise the rugged terrain within a few kilometers in the normal downwind direction, however no problems were encountered in site selections for precipitation sampling. In contrast to the washout studies performed near the heavily polluted Keystone plant, the effects of the Centralia effluent plume were isolated by rain concentration measurements. The effect of the plant on local levels of SO_2, $SO_4^=$, and H^+ was determined from laboratory analysis of the precipitation samples. The rain sampling positions were concentrated in accordance with the predominant wind direction during rainfall at Centralia. Ninety fixed collection sites were arranged to reflect the prevalent area under the plume at distances from 0.4 to 11 km downwind in the latest experiment study. A scavenging "plume" of SO_2 on the ground sampling array was clearly indicated. The measured scavenging rates for SO_2 as a function of downwind distance show rates of 3.0 and 27.0 (gram-moles/cm s) x 10^9 at 0.4-0.5 km downwind, 100 and 42 (gram/moles/cm-s) x 10^9 at 4.5 km, 54 (gram-moles/cm-s) x 10^9 at 7.0 km and 115 and 129 (gram-moles/cm-s) x 10^9 at 10-11.5 km downwind. Sulfate molar concentrations were, in general, not large compared with those of SO_2, so a deposition plume for sulfate was difficult to define, except for some discernible plume-related sulfate deposition at downwind distances greater than about

7 km. No correlations were found between locations of the power plant plume and the measured concentrations in rainwater of ammonium, nitrite, nitrate, or phosphate ions.

Along with the involved field experiments at the Centralia plant was a research program directed at the development of a numerical model for predicting precipitation scavenging of reactive pollutants from power-plant plumes. The model is described in Section IV. Comparisons of the calculated results were conducted using the model with experimental data employing the liquid phase ammonia (Scott and Hobbs 1967) mechanism. Indications were that the mechanism is insufficient to explain observed sulfate formation rates in the tested plume and raindrop situations.

The primary conclusions drawn from the Centralia field program were: (1) At downwind distances less than about 10 km, sulfate concentrations in rainwater due to the presence of the plume do not appear significant. (2) Ammonium, nitrate, and soluble (ortho) phosphate ion concentrations in the samples obtained were close to normal background levels. (3) SO_2 and hydrogen ion were the only measured species which showed plume related deposition patterns at all distances from 0.4 to 11 km where samples were collected.

In a different investigation seasonally averaged sulfate concentration in rainwater samples collected near the TVA Colbert plant were analyzed by converting the data to isopleths of sulfate washout at 17 sampling sites located in an area 1.6 to 25.7 km (1-16 miles) from the plant (Hutcheson and Hall 1974). The washout process was defined as the mass of sulfate per unit area per inch of rain. The washout pattern was found to be skewed in the general direction of maximum precipitation rates. Unlike the Keystone power plant, the Colbert plant is situated in an area of low background atmospheric sulfur concentration and has stacks only 91.5 m (300 ft) high. Little SO_2 washout compared to the sulfate aerosol washout was observed at Keystone; at Colbert, the major features of the washout pattern derived from the data cannot be accounted for by sulfate aerosol scavenging but can be accounted for if most of the washout is attributed to SO_2 scavenging. Hale's (1971) theoretical argument is supported by this study, which indicates that the background acidity of rain (with large raindrops being less acid than small drops and therefore absorbing greater quantities of SO_2), the height of the pollutant source and the rainfall rate (high precipitation rates are associated with larger mean raindrop size) all strongly influence the relative importance of sulfate aerosol and SO_2 scavenging in the very complex sulfur washout process.

Section VI

EVALUATION

Studies of SO_2 oxidation rates in power plant plumes show a diversity of results, with reported values ranging from under 1 percent per hour to as much as 30 percent per hour. The Gartrell et al. (1963) study of the coal-fired Colbert power plant plume in Alabama presented SO_2 oxidation rates of about 1-3 percent per hour for low humidity cases, while higher rates on the order of 30 percent per hour were found for higher relative humidities, with one case yielding an SO_2 oxidation rate of 55 percent during a period of 108 minutes. The Stephens and McCaldin (1971) field study of the coal-fired Crystal River power plant plume in Florida resulted in data which showed the SO_2 half-life to be 144 minutes at relative humidities ranging from 40-55 percent, and 70 minutes for relative humidities of 78-80 percent. Although there is controversy concerning the assumption made by Stephens and McCaldin that particulate matter in the submicron range can be used as a conservative tracer, their results bring out the concept of the importance of relative humidity in the plume and ambient air as a major factor in the atmospheric oxidation of SO_2. Studies of the coal-fired Keystone plant in Pennsylvania (Newman et al. 1975a) and the oil-fired Northport plant in New York (Newman et al. 1975b) yielded SO_2 oxidation rates in the coal-fired plumes that were surprisingly lower than those of the oil-fired plant. The oxidation rates ranged from 0 to 5 percent in the Keystone plumes and from 0 to 26 percent in the Northport plumes; the low particulate content of the coal-fired plume is believed to be responsible for the correspondingly low SO_2 oxidation rates. Definite correlations of oxidation with relative humidities were not evident for the experimental humidity range of 40-95 percent. At the Four Corners power plant in New Mexico (University of Utah Research Institute 1974) a recent trend of observing lower SO_2 oxidation rates in studies of coal-fired plant plumes was evident. Reported values for conversions of SO_2 to particulate sulfate ranged from 0.37 percent per hour at a distance of 21 km downwind (at relative humidities of 36-51 percent in one test), to 0.76 percent per hour at 21 km downwind, and 0.45 percent per hour at 60 km downwind, with relative humidities of 26-40 percent in another test. The dryness of the region is believed to be responsible for the low conversion rates. In the MISTT program, mapping of the Labadie plant plume near St. Louis resulted in pollutant budgets inferred from estimates of rates of transformation and removal in the plume which showed almost no sulfate formation within the first 30 km of the plant, and a conversion of some 2% hr^{-1} at downwind distances of 30-50 km.

A study of the Morgantown plant plume in Maryland by Davis et al. (1974) is controversial because of the evidence of an "ozone bulge" in the plume profile data 50 km downwind of the plant. A series of reaction steps was postulated to help interpret the bulge and there is the possibility that under certain conditions, the initiating SO_2 + OH mechanism in this reaction sequence may be the dominant SO_2 conversion mechanism to ultimately result in H_2SO_4 and other aerosol products under certain conditions. This conveniently leads into an evaluation of what the literature has shown to be the relevant SO_2 conversion mechanism in power plant plumes.

Five primary mechanisms for SO_2 oxidation in the atmosphere have been postulated. These include the homogeneous processes of direct photo-oxidation, indirect photo-oxidation, aqueous phase oxidation, the heterogeneous process involving catalyzed oxidation in liquids and dry heterogeneous reaction. Homogeneous photo-oxidation of SO_2 has generally been agreed to be an insignificant cause of sulfate formation in plumes, with the slowest rates of SO_2 conversion typically occurring in pure air and sunshine. The reported rates range from about 0.1 percent per hour in laboratory work (Gerhard and Johnstone 1958, Urone et al. 1968, Cox and Penkett 1970) to actual atmospheric observations such as a 2 percent per hour value described by Katz (1950) and one value of 12 percent per minute found by Shirai et al. (1962).

Indirect homogeneous photo-oxidation of SO_2 may be a significant reaction sequence in plume chemistry. The dominant gas phase removal mechanism for SO_2 may be the oxidation of SO_2 by OH and HO_2 as shown by Davis et al. (1974), Castleman et al. (1974), and Calvert and McQuigg (1975). A better understanding of the impact of this mechanism on SO_2 plume conversion rates will be forthcoming as more reliable measurements of OH concentrations in plumes are available. One current estimate of 2 percent per hour for SO_2 conversion through OH and H_2O oxidation has been put forth by Castleman et al. (1975). It is difficult to calculate the homogeneous rate of oxidation of SO_2 in photochemical smog systems due to the diversity of test results and complexities introduced by the use of different SO_2-NO_x-hydrocarbon and SO_2-O_3-olefin mixtures and conditions in varying experimental laboratory procedures.

Studies of homogeneous aqueous phase reactions yield SO_2 conversion rates ranging from the order of 0.1 percent hr^{-1} (Miller and dePena 1972), through 2.5 percent hr^{-1} (Scott and Hobbs 1967), to 13 percent hr^{-1} (McKay 1971) for the ammonia-SO_2-water system. Dana et al. (1976), in their field work regarding precipitation scavenging of reactive fossil-fuel plumes, place strong emphasis on the need for experimental measurements of ammonia solubility in water at low concentrations prior to any further speculation regarding the role of ammonia in SO_2 oxidation processes.

The effect of metal catalysts in the aqueous phase oxidation of SO_2 has been shown to result in a range of from 0.1 to 2.0 percent per minute, with possible higher rates. Foster (1969) estimated the rate of oxidation by a Mn catalyst to be 0.09 percent min^{-1} and 0.15-1.5 percent min^{-1} for the rate of oxidation for Fe, showing the importance of iron as a catalyst. Cheng et al. (1971) extrapolated their laboratory results to the atmosphere and arrived at an SO_2 oxidation rate of about 2 percent hr^{-1} for a $MnSO_4$ catalyst concentration believed to be typical in a power plant plume. The oxidation of SO_2 for the Cheng et al. study is seen to be four times larger than Foster's level. The high variability in oxidation rates is attributed to the sensitivity of this mechanism to parameters such as pH, relative humidity, temperature and catalyst concentration. The most effective catalysis occurs in solution under precipitation conditions; Dana et al. (1976) have pointed to faster SO_2 chemical conversion rates in plumes, with a chemistry believed to be more complex than for fair weather. The mechanisms that could be active under precipitating conditions include (1) an accelerated gas-phase reaction process, (2) a liquid phase reaction process in precipitation, (3) a liquid phase process in cloud droplets and subsequent efficient scavenging of cloud drops by precipitation, or (4) combinations of the above.

Heterogeneous solid-gas phase oxidation studies in the laboratory may not extrapolate quantitatively to the atmosphere. In power plant plumes, it is the small particles, less than 1 μm in diameter, that are potential catalysts for chemical reactions because of their large specific surface area and probably high trace metal content. A realistic estimation of the surface area available for catalytic aqueous phase oxidation or catalytic oxidation on dry surfaces requires data on particle surface composition, particle surface structure, and particle concentration in the plume; such data has not been gathered in past studies. Of interest is the role of dry particulate vanadium in the heterogeneous oxidation of SO_2; but the role of vanadium as an effective catalyst is disputable as Corn and Cheng (1972) and Urone et al. (1968) have shown it to be relatively inactive.

Researchers have presented theories which favor either homogeneous or heterogeneous mechanisms as being dominant in SO_2 oxidation in plumes, but a review of all studies suggests that each mechanism probably is of major importance but under quite different conditions. For example, the homogeneous gas phase process involving OH radicals most definitely has a strong positive temperature dependence due to the requirement of high H_2O, high O_3, and high UV flux for the generation of maximum OH concentrations (the very small negative temperature dependence of the OH + SO_2

reaction would be insignificant compared with the strong positive temperature dependence for OH production). In the case of heterogeneous processes, things are much more complex; here both positive and negative temperature dependence terms are likely to be involved. Other significant negative effects result from absorption and adsorption of gaseous species (e.g., O_3 or SO_2) on or into a liquid or solid surface. In general, it appears that in the more complex heterogeneous systems, counterbalancing temperature terms render these processes rather insensitive to temperature or that this reaction mode has a net negative temperature dependence (McKay 1971). If so, this could mean that under cool weather or wintertime conditions (also possibly summertime night conditions) heterogeneous reactions are the major SO_2 conversion mode (Davis, personal communication, 1976).

One line of thought may be that at a given power plant location, there may be no one predominant SO_2 conversion mechanism (homogeneous or heterogeneous) acting on an annual basis, as each mechanism can operate efficiently only when a specific (and as yet undefined) set of environmental parameters are present. For example, in the summer months with characteristically higher temperatures and relative humidities prevalent, the production of OH free radicals may be accelerated, thus favoring enhanced homogeneous processes of SO_2 plume oxidation in the plume. On the other hand, it may be that heterogeneous processes are less effective at higher temperatures. The entire situation may be reversed in the winter months; the entire concept is qualitatively illustrated in Figure 6a as a cyclic phenomenon with annual periodicity. On a diurnal basis, the high UV flux during daylight favors the homogeneous mechanisms, while heterogeneous reactions dominate the nighttime hours (see Figure 6b). At a given time, examination of plume SO_2 conversion mechanisms as a function of distance may show heterogeneous processes dominating close to the stack but dropping off with downwind distance. The ready availability of metal catalysts from the plume allows the heterogeneous mechanisms to operate almost immediately, whereas the homogeneous processes (specifically the

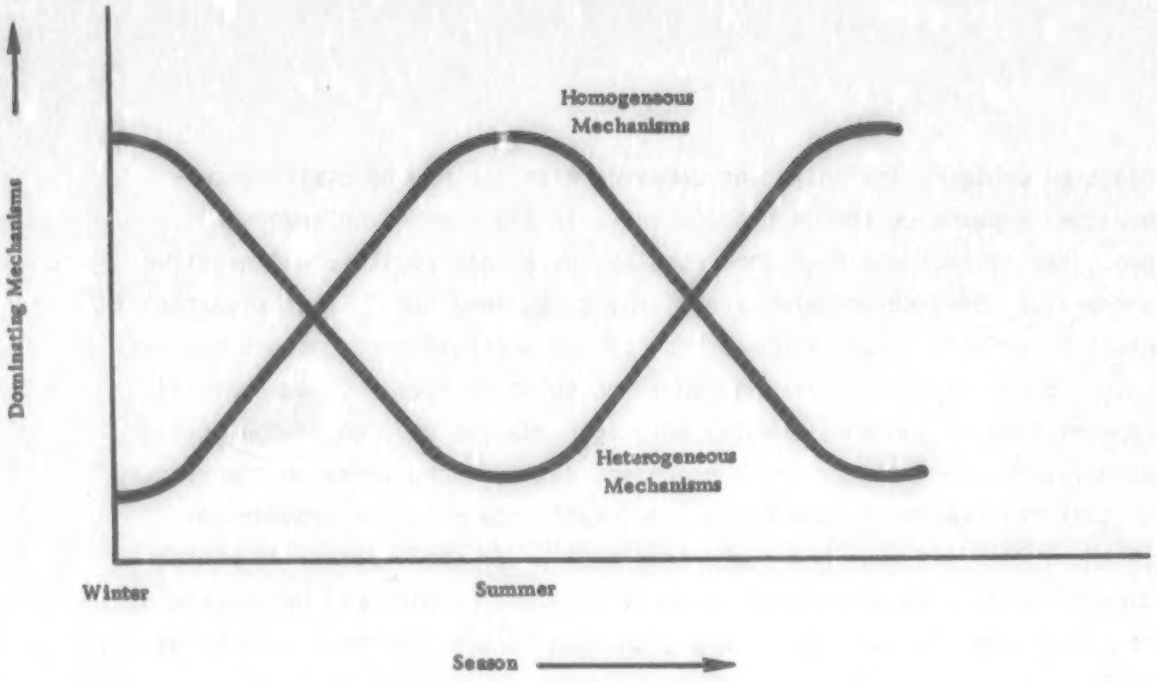

Figure 6a. Conceptual Illustration of the Variation of Both Homogeneous and Heterogeneous SO_2 Conversion Mechanisms as a Function of Time of Year for a Specific Stack Location.

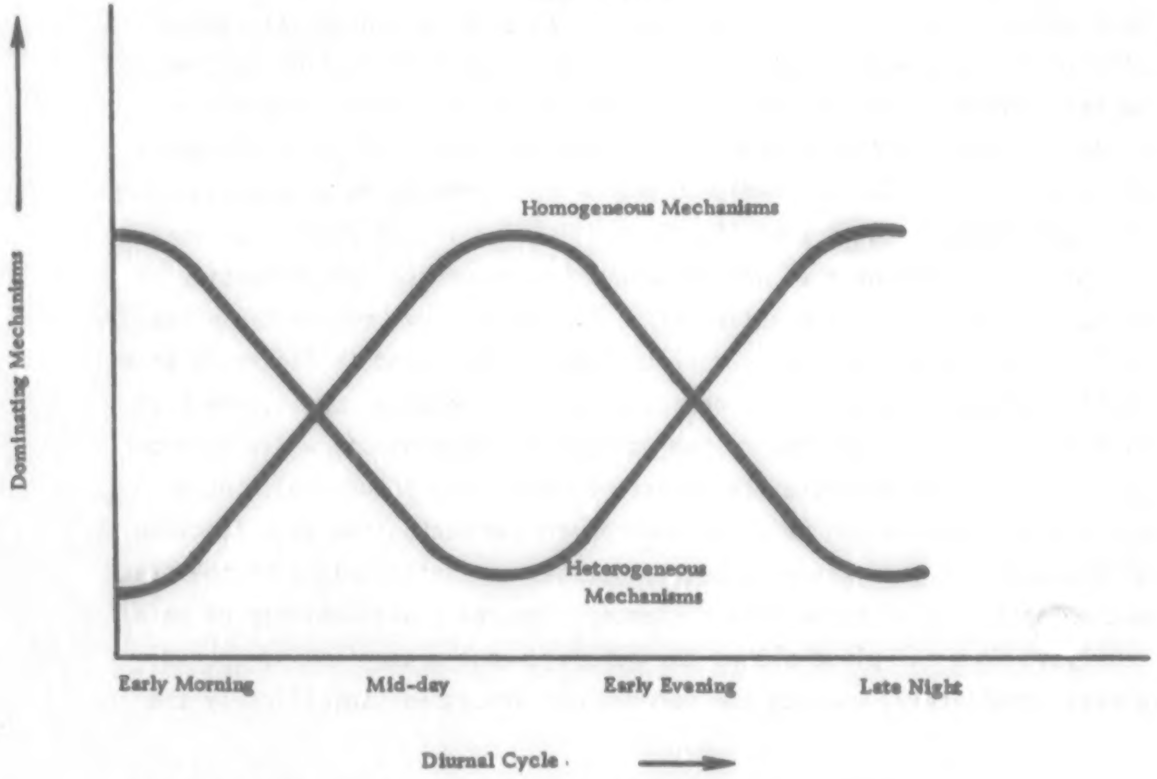

Figure 6b. Conceptual Illustration of the Variation of Both Homogeneous and Heterogeneous SO_2 Conversion Mechanisms as a Function of Time of Day for a Specific Stack Location.

SO_2 + OH reaction) take some distance downwind (10's of kilometers) to become important, possibly closely correlated with the rate of ozone production, and reaching a maximum at some point where formation is limited by dilution of SO_2 (see Figure 6c).

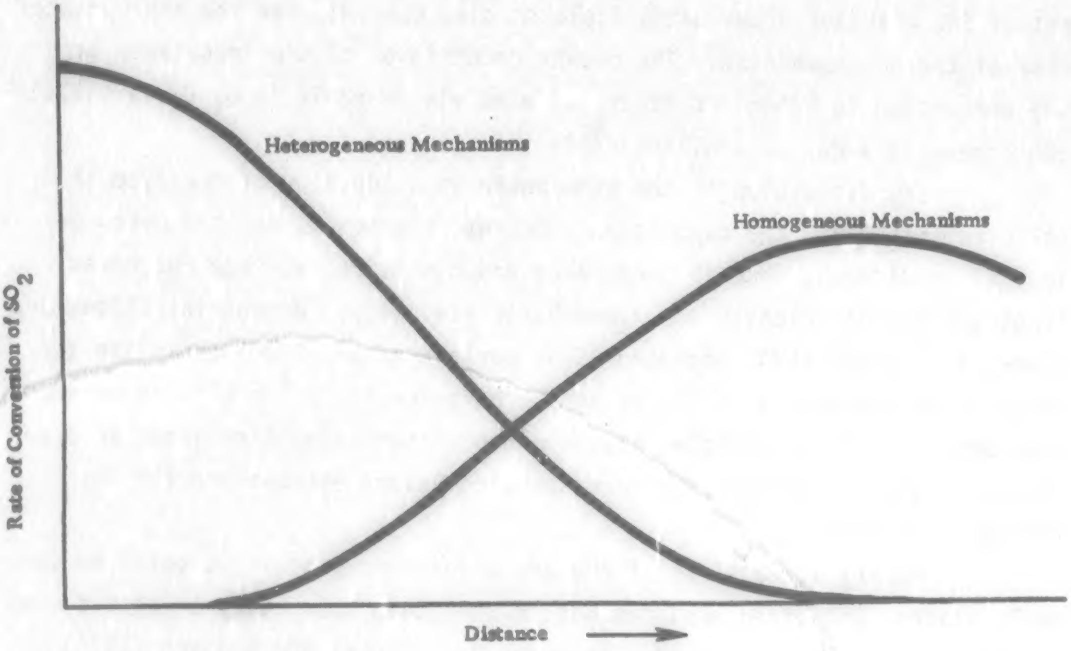

Figure 6c. Conceptual Illustration of the Variation of Both Homogeneous and Heterogeneous SO_2 Conversion Mechanisms as a Function of Distance (On the Order of 60 Kilometers) From The Stack.

What is abundantly clear is that a further quantification of the importance of the homogeneous OH-SO_2 mechanism can be determined with more complete information on OH profiles. To date, there has been only one set of OH aircraft measurements reported for the natural troposphere (Davis et al. 1976, with values ranging from 3.5 to 8 x 10^6 molecules cm^{-3} for altitudes of 7 and 11.5 km at various geographical locations). The observed OH concentrations were found to track reasonably well with the variations in atmospheric H_2O.

Chemical kinetic models to study the basic features of a reactive plume from a large point source are currently not available, but certainly will be forthcoming as quantitative information becomes available on the subjects of plume SO_2-sulfate conversion mechanisms, rates of SO_2 transformation and the extent and final state of reaction. Systems Applications, Inc. (Liu 1975) has developed one such model which employs a provisional set of SO_2 reaction steps until field studies can validate the appropriateness of the SO_2 mechanism. The recent recognition of the importance of SO_2 conversion to H_2SO_4 and other sulfates via OH radicals by Davis (1976) and others is a key accomplishment in the state-of-the-art.

Dry deposition in the atmosphere is a function of the type of pollutant material, the deposition substrate character, and the meteorological conditions, such as wind speed and direction, surface roughness length, friction velocity and atmospheric stability. Recent investigations (Owers and Powell 1974, Shepherd 1974, Garland et al. 1974) establish the velocity of deposition of SO_2 to be in the order of 10^{-2} m s^{-1}. Submicron atmospheric sulfate particles may have deposition velocities about an order of magnitude smaller than the corresponding values established for SO_2 (Chamberlain 1966).

Numerical modeling of the dry deposition process is still in the early stages, and field measurements, rather than laboratory-simulated conditions, are needed for validation purposes. Sehmel and Hodgson (1974) developed a steady-state, one-dimensional mass transfer model and with wind tunnel data, determined that for their conditions of experimentation deposition velocities were nearly independent of particle diameter only in the 0.1 to 1.0 μm size range. This corresponds to the "accumulation mode" where 80 percent of atmospheric sulfate is found. The minimum deposition velocities were found to occur in the particle diameter range of 0.04 to 0.4 μm.

Laboratory studies of the absorption of atmospheric water-soluble gases by vegetation show the rate of pollutant removal to be affected by wind velocity, canopy height, and light intensity, with the removal rate linearly increasing with increase of pollutant concentration to a limit.

The effects of the washout mechanism on power plant plume depletion are documented mostly through the research of the Battelle group in northwest Washington. The washout process depends greatly on rain characteristics such as the raindrop size distribution and the aerosol collection efficiency of the raindrops. The SO_2 rainfall deposition pattern in the region of the Centralia coal-fired plant based on rain collector samplers located on the ground along the cross-plume line clearly indicated a scavenged "plume" of SO_2 on the sampling array (Dana et al. 1976). The field experiments concluded that sulfate concentrations in rainwater due to the plume's presence do not appear to be significant at downwind distances less than 10 km; ammonium nitrate and soluble (ortho) phosphate ion concentrations were at or near normal background levels; and, the only species measured which showed plume-related deposition patterns at all distances where samples were collected (0.4-11 km) were SO_2 and hydrogen ion.

The Battelle group has also developed one of the few numerical models for predicting precipitation scavenging of reactive pollutants from power plant plumes (Dana et al. 1975, 1976). The model can predict ground level washout fluxes and average concentrations from precipitation as a function of location beneath a plume. Initial applications and comparisons of the model predictions to field data indicated that the Scott and Hobbs (1967) aqueous-phase dissolved ammonia oxidation mechanism for SO_2 is not adequate to account for observed rates of sulfate formation.

The rainout process is known to be a function of the nature of the aerosol, its size distribution, concentration, wettability, activity as a nuclei; cloud parameters such as the nature of the cloud itself, its depth, temperature and water distributions, electrical activity and other parameters. Research into this mechanism, as it applies to power plant plumes has been virtually non-existent due to the complexities involved with data gathering.

PART 3

TRANSPORT AND DIFFUSION OF PLUMES UNDER INFLUENCE OF COMPLEX TERRAIN

In contrast to the previous section, which discussed various aspects of atmospheric plume chemical transformations, this section deals with the physical and conservative behavior of plume components, i.e., with conditions in which chemical transformations do not occur (or are ignored) and in which the total mass of pollutant in the plume is treated as remaining unchanged over time. Neither of these situations is truly representative of the behavior of actual SO_2 and NO_x plumes from power plants, although both are often assumed to be sufficiently true to provide acceptable results in short-term modeling calculations of pollutant concentrations from power plant stacks.

Our ability to address directly questions of pollutant dispersion* in complex terrain is a function of our understanding of the major physical laws that govern airflow, turbulence and diffusion mechanisms under differing meteorological and topographical conditions. It is, therefore, appropriate to begin this evaluation of the state-of-the-art of plume transport and diffusion in complicated terrain with a review of the current theories and observations based upon research concerning airflow, turbulence and diffusion phenomena in such topographical surroundings. These subjects are discussed in Section VII, "Principles and Observations." Section VIII will describe the types of models available to numerically simulate these phenomena. The next section (Section IX) will be a review of field work with regard to complex terrain situations; studies concerned with the transport and diffusion aspects of actual stack plumes or tracer material will be discussed in detail, with emphasis on the program objectives, the methodologies of data sampling utilized, model-to-measurement comparisons associated with these field studies, and project results and conclusions. Included within the field studies is our own analysis of σ_y

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* Dispersion in this review is defined as the combined result of transport and diffusion processes.

and σ_z relationships in the Gaussian plume model over complex terrain as derived from LAPPES and TVA power plant plume concentration data. An overall matrix summary of these field studies concludes Section IX. Finally, Section X is an appraisal of current capabilities for calculating airflow and diffusion based upon analysis of the field studies and modeling comparisons.

Section VII

PRINCIPLES AND OBSERVATIONS

AIRFLOW IN COMPLEX TERRAIN

For the purposes of this document, consideration of airflow in complex terrain will include only studies of phenomena related to orographic features, e.g., problems of up-slope and down-slope flow. Other flows of a local nature which are not immediately relatable to differences in terrain elevation (e.g., lake and sea breezes) will not be discussed in this report. With the exceptions of mountain lee waves - only briefly touched upon in this document - and flow channeling described subsequently in discussions of field studies, the conditions described are totally influenced by diurnal effects. Daytime solar heating and nighttime radiational cooling generate the driving force for the mountain and valley flows. Whether or not these flows establish themselves is largely dependent upon the gradient conditions existing in the synoptic flows. Gradient conditions can either assist or inhibit the development of these local flows.

Several investigators have determined that a gradient wind speed of approximately 6 m sec^{-1} is necessary for the breakup of the nocturnal drainage flow. Kangos et al. (1969) have summarized much of the research that has been undertaken concerning the extent of mountain and valley winds. Maximum velocities have been found at altitudes from 1/4 to 1/2 the ridge height for upslope and downslope winds. Smith (1965) found that the depth of the drainage flow for a ridge at Twentynine Palms, California, was at least 80 m. In the same study a dome of cold air was found to develop in the valley to the west of the ridge as a result of drainage.

Egami et al. (1974) conducted a study of plume dispersion near the Mohave Generating Plant which is located in a river valley with ridges extending to 1000-1300 meters above the valley floor. Pibal measurements of wind speed and direction were taken and the summary of results concentrated heavily on the mountain and valley wind patterns. The up-valley winds were of greater depth (4000 m) than the down-valley flow (500-1000 m). The greater extent of the up-valley flow was attributed to daytime transport of momentum from higher levels due to unstable conditions.

Two wind systems have been noted by Tyson and Preston-Whyte (1972) in Natal: (1) mountain and valley winds occurring in the deep valleys and (2) mountain-plain and plain-mountain winds occurring above ridge level and above the influence of surface topography. At the time of maximum solar radiation the entire up-valley flow was as much as 1000-1250 m deep, with the maximum wind speed of about 10 m sec^{-1} at the 250-300 m level. The mountain-plain drainage wind was most fully developed at 2 hours before sunrise and became as deep as 1000 m.

The thermal circulations in a small canyon located in the Wasatch Range of northern Utah were studied by Thompson (1967). The major events of a typical night of radiational cooling were as follows: (1) formation of a thin film of cold air on the floor of the canyon and a short distance up the canyon slopes; (2) rapid wind shift from up-canyon to down-canyon wind; (3) rapidly increasing wind speed accompanying the continued cooling followed by steady wind the remainder of the night; (4) cold core persisted in the canyon as morning heating progressed and (5) rapid wind shift from down-canyon to up-canyon in the morning. The major difference between this and conditions in other larger canyons is that the inversion and nocturnal flow regime probably begin earlier and develop more rapidly in this particular small canyon.

Kangos et al. (1969) summarized work relating to air flows over mountain peaks and ridges and the resulting production of lee waves. The following summarization will confine itself only to those cases where the wind blows perpendicular to a ridge. In an attempt to classify motions over a ridge, wave characteristics such as length and amplitude were found to be important and on the basis of these, three categories were defined, namely, laminar flow, small-wave flow, and large-wave flow.

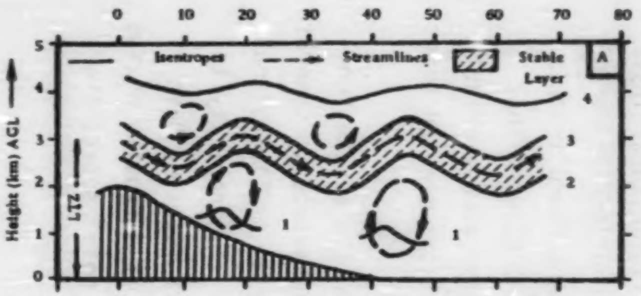
Small-wave flow is characterized by a wavelength less than or equal to the width of the ridge and by moderate winds which increase with height through a considerable depth. Stabilities are not as great as for large waves, where the wavelength exceeds the ridge width. When streamlines become packed as air passes over the ridge, large waves are produced. Rotor flow or hydraulic jump flow is usually present with large waves. Jump flow is characterized by a marked temperature inversion at some level above ridge height. The waves form a single prominent peak in the vicinity of the ridge. Large wind speeds result in the subsequent trough, and then the flow "jumps" to a higher level downstream with no further wave activity. Such a situation is depicted in Figure 7d (Lester and Fingerhut 1974). From energy conservation considerations the turbulence in this region must therefore be great.

In addition to wind speed, an important consideration in defining air flow over ridges would be the stability of the atmosphere. Those slopes facing the sun would undergo heating and hence generate upflow winds. Those slopes on the backside or receiving no sunshine could have forces acting to generate down-slope flows. The forcing equations for these up- or down-slope conditions would then need to be superimposed upon the general airflow (or synoptic flow) existing at the time.

Thus a leeward-facing slope that was receiving direct sunshine could tend to promote and enforce the rotary flow that might be generated in the lee of a mountain. Conversely, during the nighttime, or with a shaded flow the forces generating a rotary circulation would tend to be inhibited. Studies at NCAR have shown that the extremely damaging winds that occur in the Boulder area are a combination of down-slope and synoptic flows reinforcing each other.

In test releases of fluorescent particles (FP) upwind of a 300 m ridge by Smith (1965), airflows of 5 to 7 m sec^{-1} and 1 to 5 m sec^{-1} produced waves. According to theory, wind speeds this light should be characterized by flow that is displaced upward by the ridge and then returns to its previous height behind the ridge without the oscillatory pattern of well-developed lee waves. Highest concentrations of a plume should occur well above the lee slope. In the case of this particular ridge at Twentynine Palms, California, the lee slope was still quite gentle and only slight downward motions were required to bring the FP plume to the surface. The change from drainage flow to wave flow occurred abruptly as stability increased during the evening.

Lester and Fingerhut (1974) investigated the lower turbulent zone (LTZ), a highly turbulent region of near-neutral stability located immediately to the lee of a range of mountains, between the ground and the stable layer in which wave motion occurs. Aircraft flights measured potential temperature, horizontal velocity, and longitudinal gust velocity. Cross-sections of wind speed and direction and potential temperature were analyzed for a case of large amplitude lee waves and strong turbulence over the plains just east of the Colorado Rockies. The effect of the passage of a weak cold front was noted. The lee wavelength increased from 20 to 32 km, amplitude increased from 0.5 to 0.75 km, and there was an increase in overall turbulence intensity. It was not possible to isolate spatial changes in LTZ structure resulting from differences in upstream terrain since synoptic changes masked these effects. The LTZ extended 40 km downstream of the first lee wave trough. Maximum turbulence was associated with the updraft area just upstream of the rotor (see Figures 7a through 7c) and minimum turbulence was found in and near the troughs. However, in another case a stronger and more rapidly moving cold front had the opposite effect (i.e., wavelength, amplitude, depth of LTZ, and turbulent levels were all decreased). In the hydraulic jump cases (see Figure 7d),


A
Isentropes
Streamlines
Stable Layer
Height (km) AGL
LTZ
1
1
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3
4
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10
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4
5

Figure 7a. A Schematic Cross Section of the Lower Turbulence Zone (LTZ) Based On Six Case Studies, Showing the Potential Temperature Field. Evident Are The Typically Weak Potential Temperature Gradient, and the Relatively Strong Stability at the Top; The Low Level Potential Temperature Minima Are Interpreted as Evidence of Air Moving Upward From the Surface Layers In the Rotor Circulation.

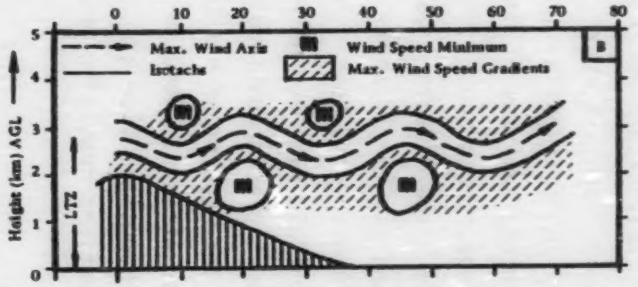
B
Max. Wind Axis
Isotachs
Wind Speed Minimum
Max. Wind Speed Gradients
m
Height (km) AGL
LTZ
0
10
20
30
40
50
60
70
80
0
1
2
3
4
5

Figure 7b. Strong Vertical Shears and Large Longitudinal Speed Changes, Especially in the Vicinity of the Updrafts and the Rotor are Associated with The Wind Speed Maximum in the Stable Layer at the Top of the LTZ.

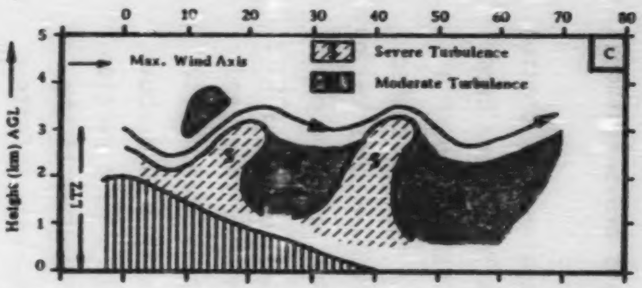
C
Max. Wind Axis
Severe Turbulence
Moderate Turbulence
S
Height (km) AGL
LTZ
0
10
20
30
40
50
60
70
80
0
1
2
3
4
5

Figure 7c. Distribution of Moderate or Greater Intensities of Turbulence Found Within the LTZ in the Wave Cases.

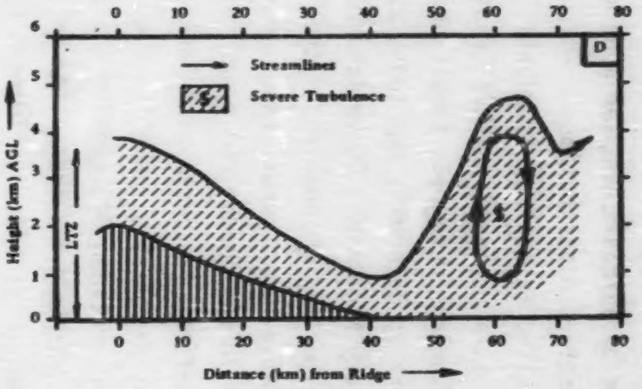
D
Streamlines
Severe Turbulence
S
Height (km) AGL
LTZ
Distance (km) from Ridge
0
10
20
30
40
50
60
70
80
0
1
2
3
4
5
6

Figure 7d. The Larger Dimensions of the LTZ and the Greater Extent of the Area Of Severe Turbulence Distinguish the Streamline and Turbulence Distribution of Hydraulic Jump Types from Wave Types.

severe turbulence was found over larger dimensions of the LTZ. In this study, the rotor circulations were deduced from examination of the streamline patterns implied by aircraft Doppler wind measurements and cross-sectional potential temperature distributions.

Channeling of airflow by mountain and valley systems was studied by Nappo (1975) as a portion of ETTEX (Eastern Tennessee Trajectory Experiment). Wind soundings were made simultaneously at five stations over a mesoscale network to determine the topographic effects on airflow caused by the Great Smoky Mountains, Cumberland Plateau, and the Tennessee Valley. It was anticipated that channeling by the Tennessee Valley would be most evident with stable night conditions. However, pronounced up-valley turning of the wind in the lowest 700 m was noted for an unstable case, and the channeling effect increased with lower altitudes. In a stable case slight up-valley channeling was noted even at approximately 1200 m above the valley floor; between 600 and 700 m a sharp change in wind direction was observed. This was the transition from the free atmosphere to the surface layer which was being influenced by the strong terrain effects. The vertical wind profiles, highly localized, showed a linear shear layer near the ground and smooth profile above, characteristic of stable conditions. Cross-valley and down-valley drainage winds were found in this lower layer. From limited analyses of the data, it was concluded that during stable conditions, large scale topographic features always affect the mesoscale flow, with the effects extending to elevations greater than 2000 m above the ground and to distances beyond 50 km.

Potential flow theory, involving use of either the stream function or velocity potential, has been extremely useful in assessing theoretical pollutant transport over mountainous terrain. The major limitations to this approach are the neglect of viscous effects and the inability to include stratification effects. Results of this type of modeling have shown that for two-dimensional flow and in the absence of diffusive mixing, ground-level concentrations resulting from an elevated point source are the same over vertically differentiated terrain as over flat terrain. The plume is closer to the surface near the top of ridges but there is also a decrease in the plume's vertical dimension (packing of streamlines) and an increase in velocity over the ridge. Streamline deformation is smaller for flow over a "hemispherical hill" than over a ridge aligned normal to the flow. Thus, there is potentially greater impact of elevated plumes on the two-dimensional hill.

For stratified flow most theoretical work has centered on solutions of the "mountain wave" equation in two dimensions. For neutral stability or for a narrow ridge, streamlines are symmetrical about the crest and the perturbations decrease with height. Egan (1975) has estimated that the effect on the ground-level concentrations at the upwind side of a ridge under such conditions would be similar to those produced by potential flow over a ridge. Streamline crests tilt upwind for stably stratified flow or for flow over broad ridges. The closest approach of a plume and the greatest velocity occur on the leeward slope. A major conclusion reached from this type of theoretical study is that streamlines approach the surface more closely under stable conditions than under neutral conditions.

TURBULENCE AND DIFFUSION ENHANCEMENT

Since turbulence is partly caused by the roughness of the surface over which air flows and since turbulence is the principal mechanism which results in the dispersion of the pollutants from a power plant stack, the dimensions of the plume, e.g., σ_y and σ_z, are frequently used to characterize the effect of terrain on the plume. Measurements of the degree of turbulence have normally been taken by means of wind observations, using bivanes to obtain both horizontal and vertical wind components or by use of tetroons to follow a particular parcel of air.

Kao et al. (1974) utilized a network of 15 continuous wind recording stations in the Salt Lake Valley to investigate typical wind patterns and the topographical effect on diffusion in valleys. The general flow patterns exhibited the expected diurnal variation with drainage winds prevailing in the early morning and evening and up-valley winds prevailing in the afternoon. Analysis of the data found that the distributions of the mean and turbulent motions in the valley were generally non-homogeneous and non-stationary. Maxima of both the mean and turbulent kinetic energy occurred in the late morning and again in the early to mid-afternoon. Alternately, trajectories of marked air parcels released successively at 10 minute intervals were computed also by Kao et al. (1974). Air trajectories of the center of mass of all marked particles released in a 3-hour period and the ensemble averages of the mean squares of the north-south and east-west components of the distance between particle trajectories for every 3-hour time period were computed for analysis of mean transport and turbulent diffusion, respectively. The mean spread of the parcels was generally greater in the afternoon than in the morning, and the east-west component of the spread (along the narrow dimension of the valley) was greater than for the north-south component. Spread was greater during the transitional evening hours than during early morning stable conditions.

Since cooling and the subsequent development of temperature inversions in valleys causes drainage of air down the slopes, inversions can be said to enhance turbulence in this situation. Leahey and Halitsky (1973) investigated turbulence at very low wind speeds at the Consolidated Edison Nuclear Plant at Indian Point, New York, located in the Hudson Valley. Wind fluctuation data were obtained from a bivane, and a turbulence climatology for winds ≤ 2 m s^{-1} was derived. The bivane observations were taken on a 30.5 m tower which was approximately 500 m from the river. The Hudson valley at this location is about 5 km wide and mountains from 150 to 350 m high enclose the valley. Diffusion was related to the turbulent wind fluctuations by the method of Hay and Pasquill (1959). Estimates were made for a distance of 1100 m from a ground-level source for all wind directions and thermal stratifications. Investigation of the data showed no occurrence of diffusion rates lower than those associated with the neutral (D) stability category.* Turbulence levels in the down-valley and down-slope flow increased as the intensity of the inversion increased, probably due to the role of inversions in producing katabatic winds.

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* For definition, see Table 11.

Leahey and Rowe (1974) completed a bivane study of airflow over a river bank near a coal-fired generating station in Alberta. Vertical distance from valley floor to rim was 52 m and the valley width was 1.5 km. The bivane was on a 30 m tower located on the valley rim. Data including four tetroon trials, each with four tetroon flights, were collected in situations ranging from very stable to near neutral. Dispersion coefficients were calculated using the mean tetroon trajectory for each trial and computing the root-mean-square of the lateral and vertical distances between individual trajectories at several downwind positions. The median values of the horizontal diffusion coefficient were twice as large as the expected flat terrain coefficient (Turner 1969) out to 3 km downwind, and about equal beyond this distance. The median values of the vertical diffusion coefficient were about twice those of Turner; the increased vertical diffusion was attributed to terrain influences.

In another study in Alberta, over Copithorn Ridge, SO_2 measurements were taken by helicopter by Leahey (1974). Copithorn Ridge is 99 m high and is located 1.2 km downwind of a stack which had an emission height of 69 m. A comparison of σ_y and σ_z during neutral conditions with the expected dimensions using a D category* indicated that the experimental results were respectively 1.7 and 1.2 times larger than would be anticipated over flat terrain, i.e., greater diffusion than expected took place.

In the study of diffusion in a deep, steep-walled canyon in Utah (Start et al. 1973), three physical mechanisms were postulated to enhance mechanical turbulence. The first effect is the downward transfer of momentum into the canyons from the considerable turbulence generated around rough mountain tops. This effect is illustrated in Figure 8 where the downward transport of momentum is related to the cross-canyon component flow. During conditions of strong temperature inversions, and with downward momentum transport from the mountain tops minimal, downslope density flows from side or feeder canyons may drain out into the main canyon and may have enough momentum to reach the opposite main canyon slope. They may then rise in a form of overshoot. Helical-like circulations may result when these feeder canyon air drainage flows act in conjunction with downslope density flows along the main canyon wall (Figure 9). The third turbulence-enhancing mechanism is wake turbulence. It is brought about by flow over and around protruding cliffs, and across small, steep-walled draws and indentations (see Figures 10a and 10b). The wake region downwind of these topographical features is characterized by a region, called a cavity, of reduced wind speed and enhanced turbulence. Wake turbulence is the smallest in scale of the three turbulence mechanisms.

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* For definition, see Table 11.

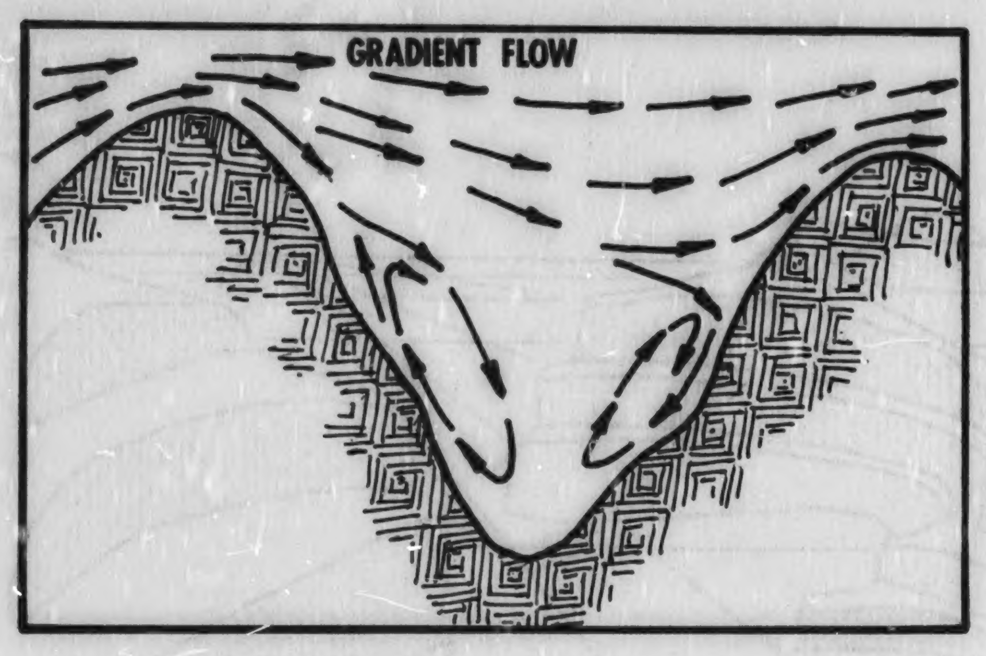


Figure 8. Schematic Illustration of Mountain Top Influences Upon the Gradient Level Flow Component and the Downward Transporting of Gradient Flow Momentum.
(from Start et al. 1973)

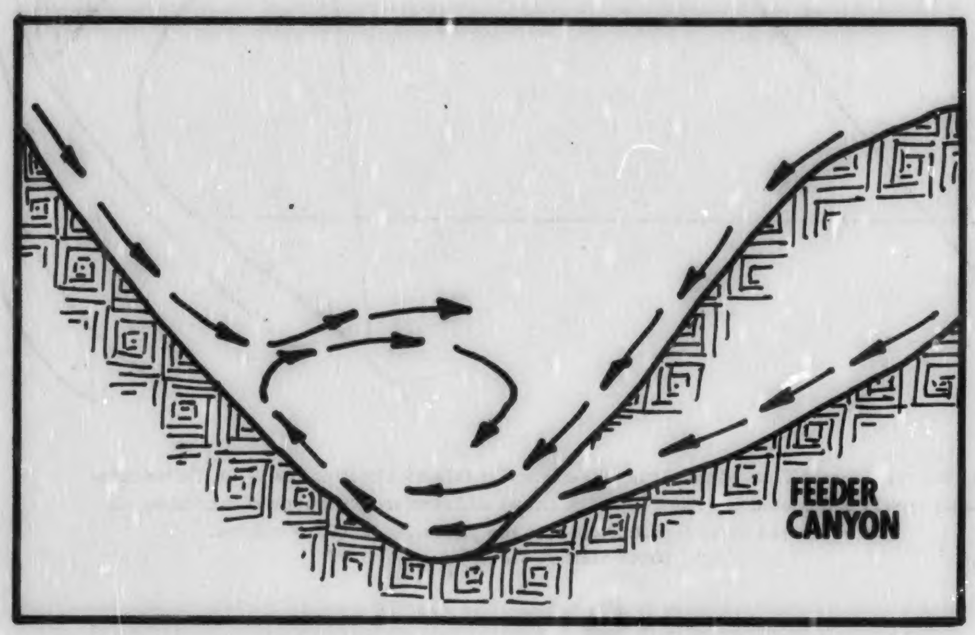


Figure 9. Schematic Illustration of Circulations Triggered by Slope Density Flows and Air Drainage from a Side Feeder Canyon.
(from Start et al. 1973)

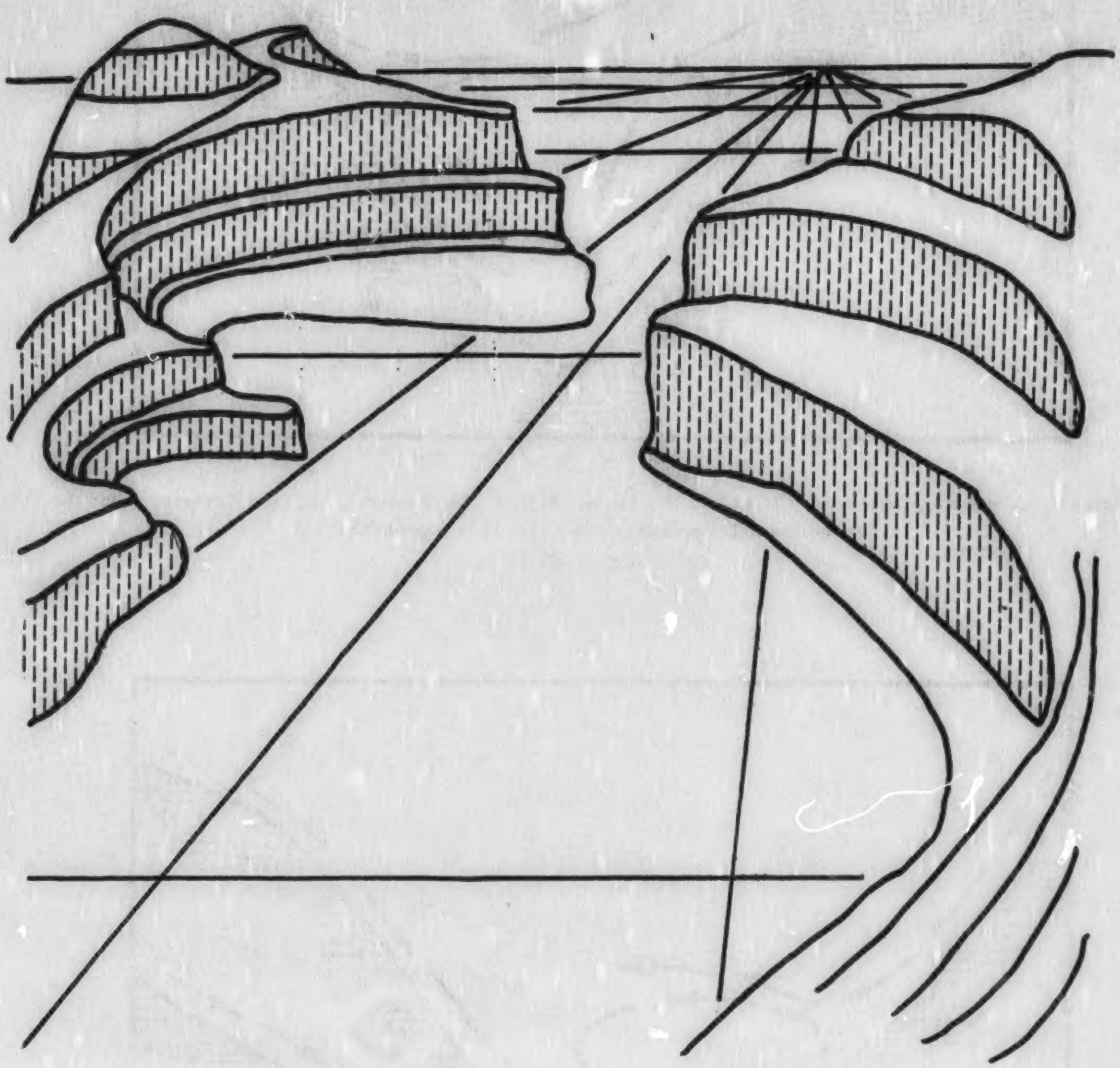
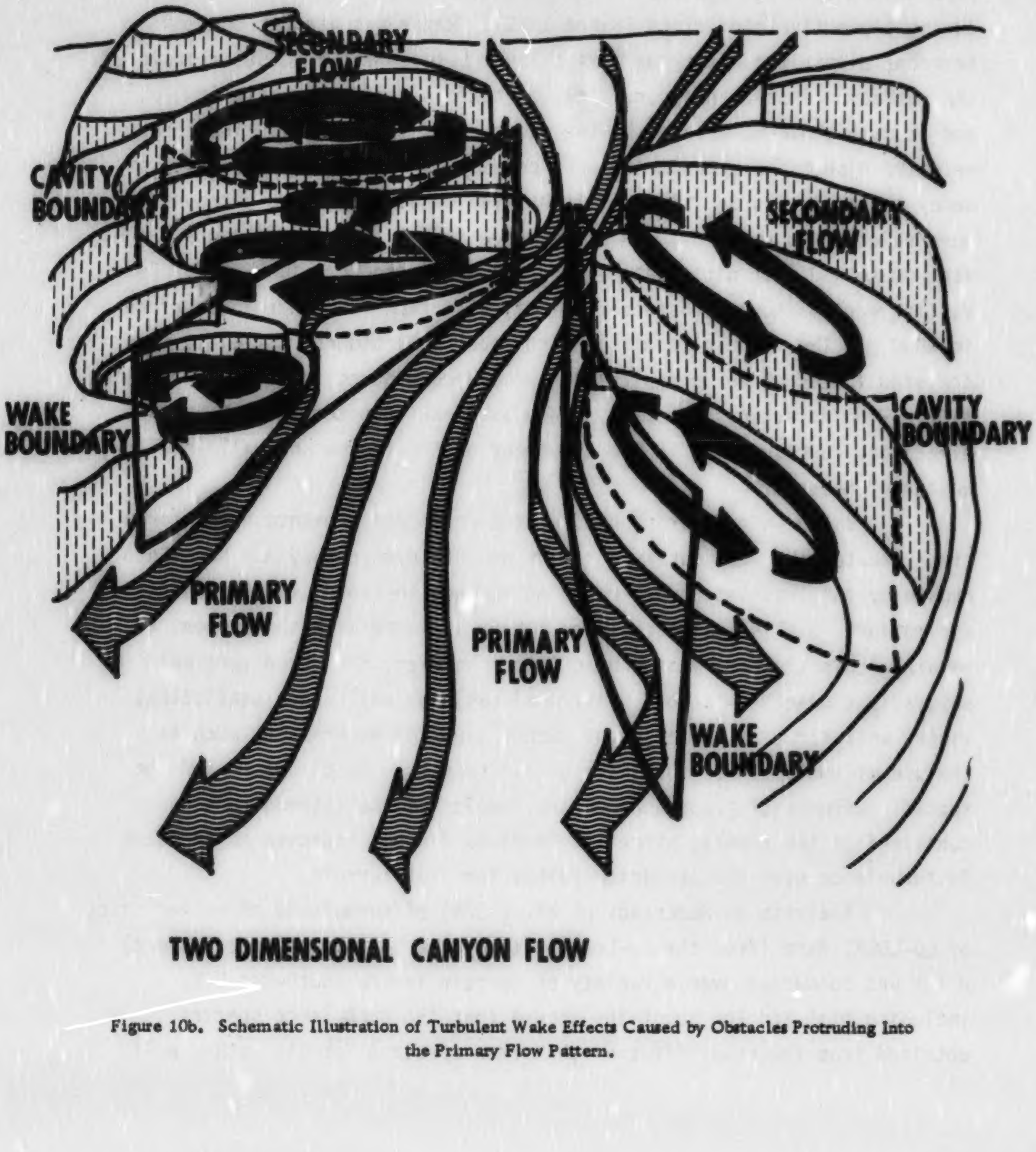


Figure 10a. Schematic View of the Type of Terrain Capable of Affecting the Wake Turbulence. This same terrain is depicted in Figure 10b with arrows added to show the type of secondary air flow caused by such protrusions into the primary flow of the canyon. (from Start et al. 1973)



SECONDARY FLOW
CAVITY BOUNDARY
SECONDARY FLOW
WAKE BOUNDARY
CAVITY BOUNDARY
PRIMARY FLOW
PRIMARY FLOW
WAKE BOUNDARY
TWO DIMENSIONAL CANYON FLOW

Figure 10b. Schematic Illustration of Turbulent Wake Effects Caused by Obstacles Protruding Into the Primary Flow Pattern.

A portion of the ETTEX project (Eastern Tennessee Trajectory Experiment) involved a study of the relative diffusion of tetroon pairs during convective conditions (Hanna 1975). Radar was used to track tetroons within the afternoon mixing layer between 500 and 1500 m above the surface. Instantaneous readings of tetroon range, elevation angle, and azimuth angle were taken by the radar every two minutes. Thirteen relative dispersion experiments were conducted near mid-day for cases of sunny or partly cloudy skies and light wind conditions. Two tetroons were released in each run; σ_z values based on the tetroon positions ranged from 60 to 430 m at distances varying between 4 and 26 km downwind. Convective eddies caused considerable vertical motion from near the surface to 2000 m. The turbulence of the wind speed (σ_w) did not correlate with the wind speed, which is typical of turbulence production due to convection. Wind directions generally had a SW or NE component, which are directions corresponding to the alignment of the ridges and valleys of eastern Tennessee.

Based on similarity theory, the predicted separation was three times greater than the observed. Part of this discrepancy may have been caused by the inability of tetroons to respond to rapid fluctuations of air motion. Statistical turbulence theory is valid if both tetroons remain within the same atmospheric layer; however, the large vertical separations detected between individual balloons may render statistical theory void for this application. Other types of experiments such as the use of smoke puffs might give better results. No discussion of the specific effects of topography on the results of the tetroon runs was contained in the report, since the findings did not indicate an increase in turbulence over the predicted values for flat terrain.

Analysis by MacCready et al. (1974) of turbulence characteristics of LO-LOCAT data (from the Lo-Low Altitude Clear Air Turbulence Project) which was conducted over a variety of terrain in the southwest U.S. including high and low mountains showed that the turbulence spectra obtained from the study fit the von Karman spectrum formula rather well, especially the -5/3 law at the small end of the spectrum. For very stable cases in complex terrain the minimum lateral turbulence observed was independent of wind speed up to about 5 m sec^{-1} and then increased with wind speed. Values of the standard deviation of vertical wind speed (σ_w) were generally 60-70 percent of the standard deviation of crosswind speed (σ_v) under stable and very stable conditions. Standard deviations of horizontal wind direction (σ_θ) were greater than "standard" σ_θ values for the stability classes as presented in AEC Safety Guide 23, at low wind speeds for all stability classes. The values for stable and very stable cases were always much greater than would have been inferred by the standard method of relating turbulence to temperature stability classifications* which is applicable for essentially flat terrain.

MacCready et al. (1974) also devised a method for relating a roughness terrain factor (σ_{tr}), obtained from U.S. Geological Survey topographic maps, to estimates of horizontal turbulence (σ_v). A regression analysis was employed to determine a best fit curve relating turbulence intensity to wind speed, height above the ground, and σ_{tr}. Results showed that turbulence values were influenced strongly by σ_{tr} and only very weakly by the former two variables. However, there was no effort made to stratify the data according to location in complex terrain (i.e., in the lee of a mountain, valley, ridge top, etc.) or by the type of meteorological conditions. It is also questionable whether the particular terrain-turbulence relation derived in MacCready's study is applicable in other areas of the U.S. in addition to the southwest.

Most studies have indicated that complex terrain contributes to alterations in air flow and to increased amounts of turbulent diffusion compared to those for flat terrain. Airborne concentration measurements appear to be the best way to observe these effects; tetroon studies have limitations in their usefulness in determining the degree of turbulence. Drainage flow and lee waves seem to be the flow characteristics most responsible for increased turbulent effects.

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* See Table 11 for category definition.

Another method of air flow investigation has been laboratory simulation, but this has proved difficult for estimating dispersion and especially over irregular terrain. This has been mainly due to the lack of an ability to achieve similarity of Reynolds Number, Richardson Number, Rayleigh Number, and other nondimensional parameters (Snyder 1972). Perhaps the largest difficulty to overcome in achieving similarity in these numbers relates to the dual nature of turbulence. For those classes of studies for which mechanical turbulence is the dominant influence, laboratory simulation has proven to be extremely beneficial and can be scaled appropriately for similarity between the simulation and the real world. For those classes of problems in which thermally induced turbulence is an important factor (which includes almost all classes of atmospheric dispersions) there does not appear to be a satisfactory means of scaling the problem for laboratory simulation. Until a relationship can be obtained for scaling thermally induced turbulence, the use of laboratory simulation for dispersion studies will be a limited application.

Section VIII

MODELING OF TERRAIN INFLUENCE ON PLUME BEHAVIOR

A large number of mathematical models, most but not all being adaptations of a Gaussian distribution concept of plume dispersion combined with a plume rise formula, have been used in concert with terrain data to calculate plume dispersion under influences of elevated and complex terrain. We begin this section with a brief description of classical Gaussian atmospheric dispersion estimates over flat terrain, and we then discuss models which have been developed to date for calculating plume dispersion from elevated point sources in complex terrain, with emphasis on their assumptions and modifications to the standard Gaussian equations.

Gaussian empirical formulas are derived from data obtained over flat terrain, utilizing the various atmospheric stability categories.* Turner's (1969) workbook adequately delineates the classical use of the empirical formulas for flat terrain situations, as recommended by Pasquill (1961) and advanced by Gifford (1961). The typical Gaussian coordinate system, containing both horizontal and vertical planes is shown in Figure 11. Equation 1 describes the concentration of gas or aerosols at x, y, z from a continuous source with an effective emission height. The basic assumption includes a Gaussian distribution in both horizontal and vertical planes of plume spread with σ_y and σ_z respectively representing horizontal and vertical standard deviations of plume concentration distribution.

$$\chi(x,y,z;H) = \frac{Q}{2\pi\sigma_y\sigma_z\bar{u}} \exp^{**}\left[-\frac{1}{2}\left(\frac{y}{\sigma_y}\right)^2\right] \tag{1}$$

$$\left\{\exp\left[-\frac{1}{2}\left(\frac{z-H}{\sigma_z}\right)^2\right] + \exp\left[-\frac{1}{2}\left(\frac{z+H}{\sigma_z}\right)^2\right]\right\}$$

**Note: $\exp - a/b = e^{-a/b}$ where e is the base of natural logarithms and is approximately equal to 2.7183.

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* See Table 11 for class definitions.

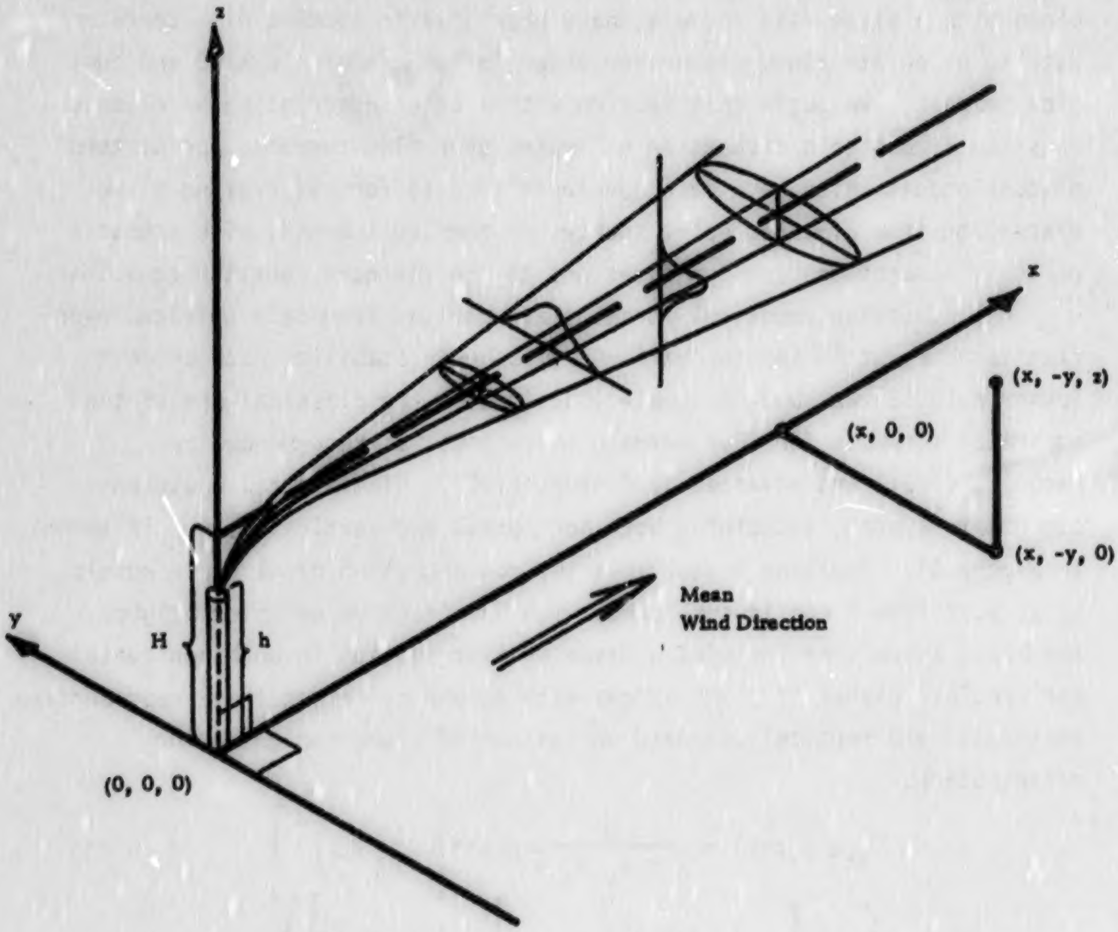
z
x
y
(x, -y, z)
(x, 0, 0)
(x, -y, 0)
Mean
Wind Direction
H
h
(0, 0, 0)

Figure 11. Coordinate System Showing Gaussian Distributions in the Horizontal and Vertical.

where

χ = concentration of effluent (g m^{-3})
Q = emission rate of effluent (g s^{-1})
$\overline{u}$ = mean wind speed affecting the plume (m s^{-1})
σ_y = standard deviation of plume concentration in the horizontal (m)
σ_z = standard deviation of plume concentration in the vertical (m)
$H = h_s + \Delta h$ = effective plume height (m)
h_s = physical stack height (m)
Δh = plume rise (m)

Several popular, similar, diffusion categorization schemes have evolved, stating relationships between the standard deviation of pollutant concentration as a function of distance and measured meteorological parameters. One of the first diffusion categorization schemes, utilized insolation, time of day, wind speed and cloud cover to define stability (see Table 11).

TABLE 11. METEOROLOGICAL STABILITY CATEGORIES (after Turner 1970)

Surface wind speed (m s^{-1})	Daytime insolation			Nighttime Conditions	
	Strong	Moderate	Slight	Thin overcast or ≥4/8 cloudiness	≤3/8 cloudiness
< 2	A	A-B	B		
2-3	A-B	B	C	E	F
3-5	B	B-C	C	D	E
5-6	C	C-D	D	D	D
>6	C	D	D	D	D

A. Extremely unstable conditions.
B. Moderately unstable conditions.
C. Slightly unstable conditions.
D. Neutral conditions.*
E. Slightly stable conditions.
F. Moderately stable conditions.

* The neutral condition D, should be assumed for overcast conditions during day or night.

The stability classification scheme as modified by Turner (1970) and defined in Table 11 has been computerized and is readily available for use on surface data. This program is available through the NOAA National Climatic Center in Asheville and is entitled STAR. The STAR program uses as input data the hourly or three-hourly data as collected at National Weather Service or FAA observing stations and the longitude and latitude of the observing station.

Another stability categorization scheme developed by TVA (1970) uses dispersion parameters for six stability classes each representing one of the following average potential temperature changes with height:

Neutral	0.00°C/100m
Slightly Stable	0.27°C/100m
Stable	0.64°C/100m
Isothermal	1.00°C/100m
Moderate Inversion	1.36°C/100m
Strong Inversion	1.73°C/100m

This categorization only considers the stable cases and is not intended for use during unstable conditions.

The NRC (formerly the AEC) uses a stability classification based upon both a temperature difference as exists close to the surface and a wind direction standard deviation (σ_θ) value as follows:

Stability Class	ΔT (°C/100m)	σ_θ
A	<-1.9	25°
B	-1.9 to -1.7	20°
C	-1.7 to -1.5	15°
D	-1.5 to -0.5	10°
E	-0.5 to +1.5	5°
F	+1.5 to +4.0	2.5°
G	>+4.0	1.7°

The present standard for estimating pollutant dispersion in the atmosphere is the Gaussian plume equation. The use of this equation implies that the dimensions of the plume can be reasonably well defined as functions of travel distance, and in terms of a σ_y and σ_z as defined in Turner's Workbook (1970) or other curves, e.g., the TVA curves (TVA 1970), obtained in previous studies. We describe subsequently, in Section IX, GEOMET's efforts to obtain a preliminary insight into the impacts of terrain in degrading the performance of the conventional Gaussian plume dispersion model. Since ground-level concentrations are often incorrectly estimated by the model, it is of interest to know how much the use of measured estimates of σ_y and σ_z improve the model agreement with measured concentrations. If significant improvement occurs, it is of interest, as a second step, to investigate the extent to which variations of measured σ_y and σ_z can be correlated with terrain features. For this analysis we needed airborne monitoring data describing plume concentrations from large power plants in complex terrain. We selected LAPPES and Kingston Plant data because previous analyses had not addressed this data from the viewpoint taken here, which is to examine whether terrain influenced dispersion within the plume. A description of our analyses, addressed toward this end, is in Section IX. Before presenting the GEOMET analysis we present a review of model modifications by other investigators and review the results by others of applying these models to available field data.

The results of several concentration measurement programs performed in complex terrain areas of both Eastern and Western United States, have led to the development of modifications to the flat-terrain diffusion model to account for the effects of terrain. Several modifications, each with differing assumptions of terrain modified wind flow and enhanced diffusion are delineated in what follows.

"NOAA" GAUSSIAN MODEL

Diffusion calculations were made in the Southwest Energy Study (Van der Hoven et al. 1972) for five planned or existing power plants located in areas of generally complex terrain (Mohave, Navajo, Four Corners, San Juan, and Huntington Canyon) with a Gaussian plume model having the following terrain-related assumptions:

- For neutral and unstable conditions, the plume centerline elevation above surrounding terrain was always equal to the initial effective plume height. Thus, the following standard Gaussian equation for ground-level concentrations beneath the plume centerline was used:

  $$\chi = \frac{Q}{\pi \bar{u} \sigma_z \sigma_y} \exp - \left\{ \frac{H^2}{2 \sigma_z^2} \right\} \quad (2)$$

  where:

  χ = short-term concentration, (applicable for periods of 10 minutes to about one hour) (gm^{-3})

- For stability classes* E and F, the plume was assumed to remain at a constant elevation above mean sea level and to impact upon surrounding terrain of equal or greater elevations. In this case, the above equation is modified so that the numerator of the term in brackets is $H-h_T$, where h_T = terrain height above stack base (m). When the terrain height exceeds H, then $H-h_T$ is set to zero.

- For fumigation conditions, centerline concentrations were estimated by

  $$\chi_{z_f} = \frac{Q}{\sqrt{2\pi}\, \bar{u} \left(\sigma_y + \frac{z_f}{8}\right) z_f} \quad (3)$$

  where

  $z_f = H - h_T$.

---

* See Table 11 for class definitions.

Maximum concentrations resulting from vertical fumigation through a layer from the terrain to the plume center were assumed to apply as follows: a critical distance, x, at which maximum concentrations caused by fumigation should be considered, was determined by $x_f = \overline{u}\, t_m$, where

$$t_m = \left(\frac{\rho c_p \frac{\partial \theta}{\partial Z}}{R}\right)(\Delta h - h_T)\left(\frac{\Delta h - h_T}{2} + h_s\right) \qquad (4)^*$$

and

$\frac{\partial \theta}{\partial z}$ = change of potential temperature with height (°C m^{-1})

c_p = specific heat of air at constant pressure (ergs g^{-1} °C^{-1})

t_m = time required for the inversion to rise from the top of the stack to the plume center (s)

ρ = ambient air density (g m^{-3})

R = net rate of sensible heating of an air column by solar radiation (ergs s^{-1} cm^{-2})

and the rest of the symbols are as defined for Equation (1).

- The following equation was applied for limited mixing conditions when the pollutant is uniformly mixed in the vertical and the receptor is below the plume centerline:

$$\chi_{z_m} = \frac{Q}{\sqrt{2\pi}\, \overline{u}\, \sigma_y z_m} \qquad (5)$$

where

z_m is the mixing depth of constant height above the terrain in meters.

---

[*] The form of equations 3 and 4 differs somewhat from the equations presented in D.B. Turner's Workbook for Atmospheric Dispersion Estimates for fumigation conditions. Equations 3 and 4 assumed fumigation conditions between ground and plume centerline, while the Workbook assumes the mixing layer to be between the ground and the plume top (where the top is defined as $2\sigma_z$ above plume centerline). As a result, centerline concentrations under fumigation conditions as estimated in the Southwest Energy Study are greater, by a factor of $1 + 2\sigma_z/z_f$, than would have been the case had they been estimated by Turner's method.

- Long-term concentrations (monthly, seasonal or annual time periods) are represented by two equations:

$$\chi = \frac{\sqrt{2\pi}\, Qf}{\bar{u}\, \theta' x \sigma_z} \exp - \left[\frac{\left(h_s + \Delta h - h_T\right)^2}{2\sigma_z^2} \right] \quad (6)$$

where

f = relative frequency with which winds blow in a given wind sector

θ' = angular width of sector (radians),

and

$$\chi_{z_m} = \frac{Qf}{\bar{u}\theta' x z_m} \quad (7)$$

for limited mixing conditions.

Equations (5) and (6) were used (Martin and Tikvart 1968) with average mixing heights, frequency distributions of wind speed and direction by stability class (generated by the "STAR" program), average terrain height profiles in each of eight sectors from the plants and assumed average plant loads of about 80 to 85 percent to calculate annual concentrations at various locations near each of the five plants.

Output of this model for short-term concentration calculations under stable conditions, with the associated terrain impaction assumption, has been compared with observed concentrations (both tracers and SO_2) more often, and for more different elevated sources in complex terrain (at least five), than has been the case with any other model. However, the comparisons in all cases have been limited to results for only a few days (< 10) at each site; further, little or no validation of the fumigation or limited mixing models has been attempted.

The consensus is that the NOAA model representation is an oversimplification of the complicated wind field-terrain interaction, and of the important terrain parameters such as the slope of the terrain and the width of raised terrain barriers in the plume path.

U.S. EPA SINGLE SOURCE (CRSTER) AND "VALLEY" MODELS

The Office of Air Quality Planning and Standards (OAQPS) of the U.S. Environmental Protection Agency has used two variations of the conventional Gaussian model for the evaluation of air quality in complex terrain (Mears 1975). The CRSTER model uses equations very similar to those presented above for the standard and limited mixing conditions in the NOAA model. The terrain adjustment procedure is the same except that it is applied for all stability conditions.* The stability class, wind speed and direction, and mixing depth can be determined through subprograms from standard U.S. Weather Service surface and upper air (radiosonde) data; these processed data can then be input to the model with effective plume height data to obtain hourly concentration values for selected receptor points which are then arithmetically averaged to obtain average values which correspond to the time scales of the various air quality standards (i.e., 3-hour, 24-hour, annual average). Although this model has been extensively used to evaluate the air quality impact of a number of power plants in complex terrain, only limited comparisons with measured concentrations have been published.

The second of OAQPS's complex terrain models is called "VALLEY" and is used for a worst case 24-hour average prediction of plume impact for cases where the plume centerline is below the elevation of receptors located on nearby terrain. It also utilizes a Gaussian formula very similar to that of the NOAA model for stable conditions (i.e., with the same basic terrain adjustment formula) with the following differences:

- The height of the plume centerline above the ground is decreased by the elevation of the ground above the base of the stack.

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* See Table 11 for stability definitions.

- The effluent is uniformly distributed in the horizontal over a 22.5° sector for estimates of crosswind dispersion.
- The plume centerline is never less than 10 meters from the ground.
- The wind direction is within a given 22.5° sector for 6 hours out of 24.
- Constant 2.5 m/s winds and E stability class are assumed throughout the 24-hour period.

ERT PSDM MODEL

Environmental Research and Technology, Inc. (Egan 1975) has developed a modified Gaussian model for application to point sources in complex terrain, for neutral, unstable and stable conditions with assumptions as follows:

- For receptors located on terrain above the effective plume height, the model assumes the plume to remain at a distance generally taken to be one-half of the effective plume height over flat terrain away from the surface, for neutral and unstable conditions.
- For stable conditions the plume is assumed to pass around the side of a hill rather than over it.

There is no evidence of validation of this model in the literature.

Egan (1975) has made further modifications to the Gaussian model based on results of laboratory analyses of flow past a cylinder and sphere for neutral conditions. The modified ground-level centerline plume equation, including reflection at the surface, is

$$\chi = \frac{2Q}{2\pi\ \sigma_y \sigma_z\ \overline{u}\ D_y D_z} \exp\left[-\frac{1}{2} \left(\frac{\eta H}{\sigma_z\ \zeta\ D_z} \right)^2 \right] \qquad (8)$$

where

D_y, D_z = Coefficients used to modify the σ_y and σ_z values due to the effects of complex terrain. (i.e., D_y and D_z would equal one for flat terrain)

ζ = $(\partial\psi/\partial H)/(\partial\psi/\partial z)$, the ratio of the vertical gradient of the stream function at the effective stack height to the gradient at the plume centerline over the surface (dimensionless). The ratio reduces to one for flat terrain.

η = $(z - z_o)/H$, where z (m) is the local height of the plume, z_o (m) is the local height of the terrain, and H(m) is the effective height of the stack. This ratio reduces to one for flat terrain.

This model constitutes a departure from the conventional Gaussian formula in that it (1) accounts for the alteration of flow fields by complex terrain (through introduction of the parameter "ζ"), and (2) allows a conceptual separation of the effects of complex terrain on the flow field and turbulent diffusivities. Unfortunately, these changes require estimates (either from a flow model or from measurement studies) of the relative height above the surface of the plume centerline (a function of terrain), and the streamline spacing in the vicinity of the plume in order to be applied effectively. This requires a more extensive meteorological data base (at least with respect to spatial resolution) than is normally available for estimating diffusion in complex terrain with the standard Gaussian models.

AEROVIRONMENT STATISTICAL TURBULENCE MODEL

MacCready et al. (1974) have developed an interesting model in which they have used classical turbulence-diffusion theory together with turbulence spectra determined from the Air Force - Boeing LO-LOCAT program conducted in areas of mountainous terrain of the southwest to develop a modified Gaussian model for use in complex terrain.

The relationship between turbulence and diffusion is developed starting with Pasquill's (1974) theoretical equation describing expanding crosswind dispersion of a puff in an isotropic homogeneous environment, given by

$$\sigma_y^{2} = \overline{v'^{2}}t^{2}\int_{0}^{\infty} F_e(n)\left[\frac{\sin \pi nt/\beta}{\pi nt/\beta}\right]^{2} dn \tag{9}$$

where

σ_y is the crosswind dispersion coefficient (m)

v' is the turbulent velocity in the direction crosswind to the mean wind direction with the averaging referring to an infinite ensemble average (m s^{-1})

t is the time of travel (s)

β is the Lagrangian* correlation coefficient (dimensionless), and

$F_e(n)$ dn is the Eulerian** power spectral density in the frequency width dn.

The averaging refers to an infinite ensemble average, and material is released at time t = 0. MacCready et al. (1974) states that Taylor's hypothesis which converts temporal to spatial frequency, together with empirical estimates of β and the von Karman turbulence spectra (which the LO-LOCAT data were shown to represent well) were then used to arrive at the formulas

$$\sigma_y^2 = 0.44\ \sigma_v Lt \tag{10}$$

$$\sigma_z^2 = 0.44\ \sigma_w Lt \tag{11}$$

where

L is the Eulerian longitudinal length scale (m)

σ_v and σ_w are Eulerian lateral and vertical components of turbulence, respectively (m s^{-1}).

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* The concentration statistics are described in terms of the statistical properties of the displacements of groups of particles released in a fluid.

** Statistics are formulated in terms of the statistical properties of the velocities measured at fixed points in the fluid.

Substitution of (10) and (11) into the basic Gaussian formula gives

$$\chi/Q = \frac{1}{2.76L\ x\sqrt{\sigma_v \sigma_w}} \qquad (12)$$

for centerline concentrations, for stack height = 0 and continuous emissions.

The above equation was deemed to be applicable for one hour concentration averages, and does not take into account ground reflection or vertical shear. Regarding the latter, the authors suggest use of a $\sigma_{v_{eff}}$ rather than σ_v given by

$$\sigma_{v_{eff}} = 0.07\ u + 1.2\ \sigma_{v_1} \qquad (13)$$

where

σ_{v_1} is the measured lateral turbulence (m s^{-1}).

The validity of this assumption has not been tested; the authors suggest confirmation from additional turbulence data when they become available. The principal drawbacks of this approach are:

- Turbulence statistics σ_v and σ_w and the length scale L must either be measured at the site for which concentration estimates are desired (at a point of sufficient height to be at least midway between the ground and effective stack height) or estimated from other data.
- It has not been successfully validated.

The authors have suggested a method whereby σ_v can be estimated from a terrain roughness factor σ_{tR}, which is found by spectrum analysis of terrain height values in the vicinity of the source in question. Once

σ_{tR} is known, the relationship between σ_v and σ_{tR} derived from LO-LOCAT data can be used to derive σ_v. The following objective method is suggested for determining σ_{tR}:

(1) On a 1:250,000 USGS map plot four symmetrical lines of 45 nautical mile length in a tic-tac-toe pattern around the site of interest, with the middle of each line coming to 10 miles from the center point,

(2) Take height readings at 0.5 mile intervals for each line,

(3) For each 7.5 mile segment on a line calculate the root-mean-square about the mean height (call it $\sigma_{7.5}$), and

(4) Find the average of all the $\sigma_{7.5}$ for all segments on all lines, and multiply by 2.3 to give σ_{tR}.

This method was derived by taking root-mean-square values for segments of various lengths for the legs of various LO-LOCAT flights over rough terrain, and seeing what segment length appeared to work best and what multiplication factor gave the best fit.

Supporting the validity of this objective method is the fact that the data on which it is based (LO-LOCAT) are extensive, consisting of 8,800 turbulent spectra determined for complex terrain of varying degrees (i.e., high mountains, low mountains) and in different geographic areas (California, Colorado, New York). The spectra were also determined for all seasons and times of day. The chief drawbacks are that (1) the spectra are representative of only two altitudes, 76 m and 229 m above ground; (2) the relationship between σ_v and σ_{tR} show a large degree of scatter, and (3) σ_w must be inferred from σ_v based on the correlation between the two.

INTERA MODEL

INTERA Environmental Consultants (Hoffnagle et al. 1975) have recently developed a combined potential flow-turbulent diffusion model for use over complex terrain. The model consists of the modified Navier-Stokes equations with some basic assumptions. These assumptions are that pressure changes do not affect density; density changes have a negligible effect on viscous terms of the equations; turbulent effects are expressed in terms of eddy viscosity ($\nabla\mu$); and density can be presented as a function of position. Turbulence is expressed in terms of the pressure equation as:

$$\nabla^2 p = -\left[\frac{\partial}{\partial x}\left(\rho\frac{du}{dt}\right) + \frac{\partial}{\partial y}\left(\rho\frac{dv}{dt}\right) + \frac{\partial}{\partial z}\left(\rho\frac{dw}{dt}\right)\right] - \left[\frac{\partial}{\partial x}\nabla\mu\nabla u + \frac{\partial}{\partial z}\nabla\mu\nabla v + \frac{\partial}{\partial z}\nabla\mu\nabla w\right].$$

where the first bracketed term represents the inertial sources and the second bracketed term represents the viscous sources (a complex tensor function of the flow field).

As far as the boundary conditions are concerned, the z direction is bounded by the ground surface with variable elevation, and by the top of the turbulent boundary layer. It is assumed that

$$\left.\begin{array}{c} u = f\ (y,z)\ \text{(known)} \\ v = w = 0 \end{array}\right\}\ \text{at } x = 0$$

The y boundaries and the top of the turbulent boundary layer are assumed to be frictionless, and $u = v = w = o$ at the ground. A vertical pressure gradient is specified at the outflow face. Viscosity is calculated from the measurement of shear stress at the top of the turbulent boundary layer. For the detailed equations, the reader is referred to "Evaluation of Selected Air Pollution Dispersion Models Applicable to Complex Terrain" (INTERA 1975).

The INTERA model computes a terrain induced wind field which is then used in the diffusion equation solution, thus avoiding the parallel or horizontal flow assumptions made in the NOAA and EPA Gaussian models. This model has been rather extensively compared with observed concentrations of either SO_2 or tracer resulting from elevated point source releases at several different sites in complex terrain, where power plants or smelters currently exist (i.e., Navajo Generating Station, Garfield Smelter), or have been proposed (Kaiparowits Power Plant). The results of these comparisons are described in Section IX, for the respective study sites. The limitations in the use of the INTERA model are that the model, as it currently exists, prevents the formation of recirculating flow in the lee of an obstacle (i.e., flows as depicted in Figure 7d) and that the elimination of consideration of the energy balance equation prevents calculation of natural convective flows such as mountain/valley flows and lake and sea breezes.

TVA MODEL

The TVA model (Montgomery et al. 1973) is actually a set of Gaussian calculational techniques very similar to the NOAA model, with each technique being appropriate to a different meteorological condition. It is not specifically designed for application in complex terrain situations. The model differs from the other standard Gaussian models (EPA "VALLEY," NOAA) in that the diffusion coefficients used (σ_y and σ_z) are not the same as the standard coefficients (Turner 1969), and were essentially derived from observations of power plant plumes.

The diffusion coefficients obtained were primarily derived for the stable atmospheric conditions of limited mixing and inversion breakup. It is interesting to note that one recent study (Dames and Moore 1976) compared the NOAA and TVA coefficients using Denver, Colorado data. The NOAA model identified inversion breakup as a potential problem in meeting Colorado standards and showed limited mixing as being no problem. Using the TVA approach, just the opposite was found, i.e., limited mixing was the problem and inversion breakup was not.

Section IX

FIELD STUDIES

Field studies concerned with the physical behavior (i.e., transport and/or diffusion) of actual stack plumes or tracer material in complex terrain will be discussed in this section. Many of the 12 study locations discussed deal with tracer releases and not with actual effluents. The text will first describe some of the early fluorescent particle (FP) diffusion trials in complex terrain (McMullen and Perkins 1963, Hinds 1970), and discuss the turbulence measurements near the Dickerson Power Plant in Maryland (Weil 1974). Programs at the Naughton Plant (Spangler et al. 1973), the Four Corners Plant (Niemann 1973, Smith and Anderson 1974), the Navajo Plant (Hovind et al. 1973b, NAWC 1974, Rockwell, MRI and SAI 1975), the Huntington Canyon (Hovind et al. 1973a, c, 1974, Start et al. 1973, 1975), the Garfield Smelter (Start et al. 1974) and Kaiparowits (NAWC 1974) are presented next, followed by descriptions and GEOMET analyses of LAPPES program data (Schiermeier 1970, 1972) and data from the TVA Kingston Plant.

DUGWAY, UTAH

Some of the earliest FP diffusion trials conducted in complex terrain involved four instantaneous elevated line source releases over mountainous terrain near Dugway, Utah (McMullen and Perkins 1963). The site chosen for the trials provided successively, in a west to east direction, a large flat area at an elevation of approximately 1300 m MSL, a ridge rising to 2000 m MSL, followed by a broad valley. Elevated line source releases were carried out from 305 to 670 m above the terrain. The objective of the trials was to determine the degree to which an FP tracer cloud disseminated above a ground inversion enters into the drainage wind system characteristic of nighttime flow in a canyon-like complex terrain environment. The orientations of all of the elevated cross-wind line releases were approximately perpendicular to the main north-south ridge of the mountain system (see Figure 12). Three of the four trials were characterized by moderate to strong low level inversions, while high winds prevented well-established drainage conditions during the other trial. The wind direction was predominantly south-southeast during each of the trials, and thus lent similarity to the roughness fetches in each case.

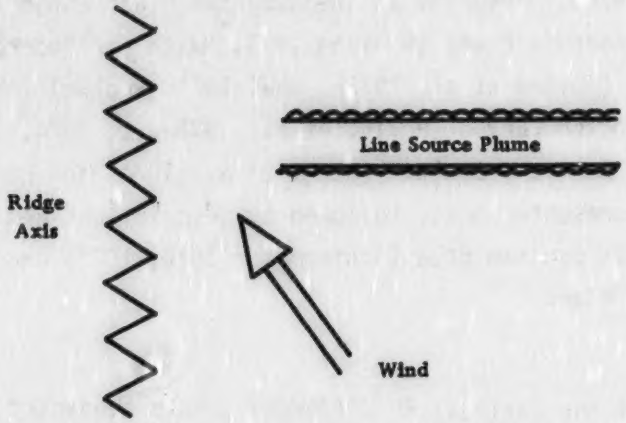

Figure 12. Configuration of Elevated Line Source Releases with Respect to Wind Direction and Orientation of Mountain System Near Dugway, Utah.

Larger amounts of tracer particles were recovered by Rotorod samplers at ground level over the rough terrain than over the flat terrain, thereby indicating enhanced rates of mixing in the irregular terrain. This was further substantiated by observing that over rough terrain FP was recovered much closer to the source. McMullen and Perkins suggest that two separate diffusion regimes contributed to the total tracer recoveries: direct diffusion from the main cloud as it passed overhead, and diffusion within the drainage flow. The drainage transport would lag in time the detection of the initial plume. A sampler would first detect the passage of the main plume as it went overhead; then, at some later time, the flows that were coming out of the canyons which had accumulated some FP material would start to show up on the sampler as their drainage flows would pass over the sampler. The percentage of FP material recovery was determined by first knowing the mean efficiency of the Rotorod samplers for the FP material used; then calculating the effective flow rate and dividing this flow rate into the reported recovery, thus giving the corresponding dosage. Direct diffusion was estimated to account for 85 percent of the recovery for non-canyon samples, however, drainage transport was estimated to account for as much as 50 percent of the recovery at samples within the canyon. The plume was definitely ill-defined, as there was no systematic variation in the total recovery with sampler location in the canyon.

McMullen and Perkins found that terrain-induced mechanical turbulence was sufficient to give significant FP recoveries at the surface in all trials both in the lateral canyons and over the open sampling area, despite low-level stability. They also found that vertical mixing is much more rapid over complex terrain than for an equal distance of travel over flat terrain. The authors suggest that the second conclusion would probably not be true for FP particles released with the flow perpendicular to the ridges instead of parallel to them.

VANDENBERG AIR FORCE BASE

Hinds (1970) reported on a series of 113 diffusion tests over the mountainous terrain in the southern portion of Vandenberg Air Force Base in Southern California. The releases were from ground-level point sources and consisted of 15-30 minute releases of zinc sulfide. Elevations in the area varied from 30-500 m AMSL. A comparison of diffusion over mountainous terrain and flat terrain in the same maritime climate showed that daytime unstable conditions minimized the importance of terrain, whereas nighttime stable conditions especially in winter led to apparently significant interactions between terrain-induced drainage flow and synoptic-scale onshore gradient flow in the complex terrain. Figure 13, from the Hinds study, shows this comparison. The solid line represents moderately unstable conditions (Stability B) as suggested by Pasquill (1962).

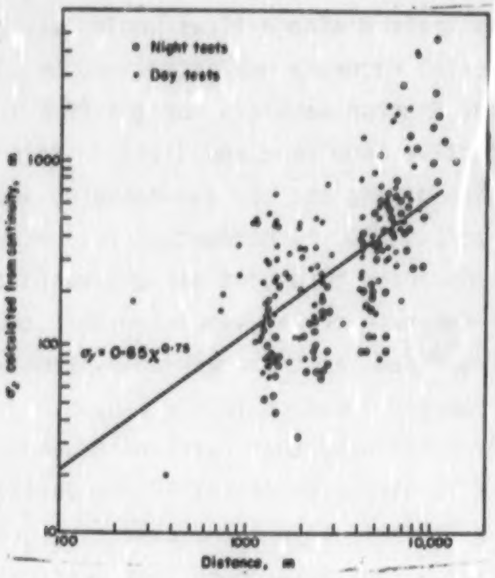

Figure 13. Vertical Plume Dimensions (σ_z) Inferred from Cross Width (σ_y) and Surface Centerline Concentration Assuming 50 Percent Deposition at the Surface (Hinds 1970).

Hinds only briefly related these phenomena to the observed concentration levels. He pointed out that on clear, winter nights with onshore pressure gradient winds it took a relatively strong pressure gradient to overcome the drainage flows in the south Vandenberg region during the existing radiation cooling conditions. Thus, onshore night wind in south Vandenberg, associated usually with strong pressure gradients and a neutral atmospheric condition, resulted in relatively low exposures inland from the source of release. No explanation is given for this other than the note that the dispersion is good during these meteorological conditions, hence there is a lower concentration downwind.

Multiple regression analysis was used in an attempt to develop relationships between meteorological parameters and normalized concentrations.

The variables chosen were travel time, standard deviation of wind direction (σ_θ), temperature lapse rate, and wind speed. Downwind travel time was found to be a relatively good predictor, but σ_θ was the poorest indicator of dispersion. The lack of dependence on σ_θ was attributed to the nonhomogeneity of the flow field in complex terrain. Impingement of the plume was probably great on the ridges, as the canyon exposures were typically only half the concentrations found on the ridge tops.

DICKERSON POWER PLANT

Increased turbulence due to rough terrain was clearly demonstrated at the Dickerson Power Plant along the Potomac River in Maryland. The study for the Maryland Power Plant Siting Program (Weil 1974) involved comparison of stability classes from Dulles Airport, which is located on flat terrain, and from the Dickerson site in moderately rough terrain, 30 km distant. The terrain in the vicinity of the Dickerson plant is characterized by generally rolling terrain but with mountains located 8 km west of the plant (Catoctin Mountains) with elevations above the plant stack top and which could equal or exceed the effective plume height on occasion. The stack base is situated approximately 90 m above MSL. The topography rises to approximately 65 m above the stack top at 10-15 km to the east of the plant. One hundred twenty-eight crosswind traverses were made with a mobile van during which the SO_2 plume from the Dickerson Plant was encountered. Sampling from the van was supplemented by pibal releases during the sampling period, by temperature and wind profiles measured two or three times a day with radiosondes and instrumented aircraft, and by wind and lateral turbulence measurements at two levels (10 and 100 meters) on an instrumented tower near the plant.

Stability classes based on Dulles data were computed every 3 hours for June 1973, and for each such stability class the standard deviation of the fluctuation of horizontal wind direction (σ_θ) was computed from lower and upper (10 m and 100 m) tower winds at Dickerson for three 10-minute periods centered around the Dulles reading. Wind direction observations were taken every 10 seconds at the Dickerson tower. A classification system was developed (Slade 1960) with a σ_θ value of 25° assumed to be equivalent to the A stability class and with σ_θ values ranging down to 2.5° for F stability. This system was then applied to both the 10 m and 100 m levels. For the 10 m level the median stability class using Dickerson data for each 30-minute period were tabulated against the stability class using Dulles data. Low-level turbulence was greater at Dickerson than at Dulles, as the medians for Dickerson were more often less stable than the respective stability classes for Dulles. However, the Dickerson 100 m level tended to be more stable than Dulles. This was to be expected since Dulles represents low-level turbulence over flat land. The Dickerson topography caused extra low-level turbulence, but this mechanical turbulence was dissipated by the time it reached 100 m.

Using the standard Gaussian formula without terrain adjustment (i.e., assuming the plume remains at a height equal to the initial effective stack height over complex terrain), and with the assumption that stability classes determined from weather observations at nearby Dulles Airport are applicable in the area of the plant, calculated concentrations stated by the authors to under-estimate 128 measured five minute ground level SO_2 centerline concentrations by as much as two orders of magnitude. The model was then rerun using the assumptions that (1) the plume remained at a constant height above MSL regardless of location (this was applied for all stability classes, as for the EPA single-source CRSTER model), and (2) the presence of rougher terrain effectively shifted the stability class by one class toward more unstable conditions. The modification resulted in an improved relationship between calculated and measured values, the former being within a factor of two of the latter most of the time.

NAUGHTON PLANT

North American Weather Consultants (Spangler et al. 1973) conducted a six-day plume tracking experiment in the vicinity of the Naughton Plant (owned by Utah Power and Light) near Kemmerer, Wyoming, under "a variety" of meteorological conditions. Their initial draft summary report pertained mainly to results obtained during one day (March 25, 1973), on which an extremely stable air mass dominated the area, and the wind was blowing towards the three surface SO_2 monitors. The study had three objectives: (1) to measure the initial plume dilution during the momentum-dominated phase of plume rise, (2) to measure the magnitude of the plume centerline concentration downwind, and (3) to relate plume measurements to ground concentration measurements from three existing monitoring stations. Along with SO_2 measurements, aerial temperature profiles were taken; one just upwind of the Naughton Plant and the other 4.8 km downwind. Surface wind observations were made at the three SO_2 monitoring sites, and two pibals were launched at mid-morning on March 25. The temperature profiles showed the very strong inversion in the lowest 470 m, and the winds at the plume level were 4.8 m/s and 4.6 m/s from the west for the two pibals launched. This led the researchers to classify the morning as a class F stability case.

In trying to discover the amount of initial plume dilution after the momentum-dominated phase of plume rise, a helicopter penetration into the plume, in an upwind direction (toward the stacks) done at various angles and altitudes, was used. The results indicate that the calculated concentration directly above the stack centerline was higher than the measured values. Out of 8 dilution factors calculated from measurements on various days, 5 were in the 45-50 range, with the full range being 23-60. No pattern of dilution factor versus stability condition was immediately obvious. On the morning of March 25 (the extremely stable case), the initial stack concentration was calculated to be 211 ppm SO_2. Initial dilution after the momentum-dominated phase of plume rise was measured at 4.90 ppm at the plume centerline. In this case, the reduction factor was 43.

The second objective of the study was to determine the plume centerline concentration downwind. The centerline of the plume was located and measured with the helicopter by flying downwind and "wagging" (side to side) and "porpoising" (up and down) to stay on the centerline of the plume. The centerline SO_2 concentration results obtained during the March 25 case are presented below. At 4.8 km and 24.1 km downwind of the plant, cross-wind traverses were flown at incremental altitudes. At 4.8 km the range of the maximum concentration at the different levels ranged from 0.08 ppm at 2300 m MSL to 1.06 ppm at 2450 m MSL to 0.07 ppm at 2600 m MSL. At 24.1 km the range was from 0.11 ppm at 2350 m MSL to 0.59 ppm at 2450 m MSL to 0.07 ppm at 2500 m MSL.* No traverses were made below 2350 m MSL or above 2500 m MSL 24.1 km downwind of the plant. A "porpoising" maneuver between 2400 m MSL and 2550 m MSL 14.5 km downwind from the plant gave a range of SO_2 concentrations of 0.05 ppm at 2400 m MSL to 0.78 ppm at 2450 m MSL to 0.06 ppm at 2550 m MSL. These measured results were compared with the SO_2 concentrations predicted by the Gaussian distributions used in the NOAA model (Turner 1969).

The comparisons were for one day during which the area was under the influence of a high pressure system, with accompanying very stable lapse rate conditions. Further, the comparison was made only for measured values obtained in the plume at distances out to about 20 km east of the plant, where terrain features are much less rugged than the mountainous areas to the west of the plant. The results of the comparison were similar to those obtained under similar stability conditions at the Four Corners and Navajo Plants, specifically, greater model overestimation of concentrations occurred at distances close to the plant than at greater distances. The NOAA model SO_2 centerline concentrations were greater than 7 times those observed at distances within 2 km of the Naughton plant, but were within a factor of 2 at distances beyond 7 km. As for the comparisons made at the Four Corners and Navajo plants, the failure of the model to adequately take into account the initial dilution (a factor probably not related to surrounding terrain) was also the cause of the model-to-measurement discrepancies close to the plant.

The third objective of the study was to relate plume measurements to ground concentration measurements from three existing air quality monitoring stations located at about 2100 m MSL. Data from the ground concentration measurements at the three existing air quality monitoring stations were only reported for the very stable case on March 25, 1973. The plume was observed to clear all terrain by a minimum of 120 m. No significant concentrations were recorded in the morning, but after destabilization of the lower atmosphere up to the height of the plume at 1045 MST (marked by a sudden decrease in elevated centerline concentrations), concentrations slightly above the background concentrations were recorded. The maximum concentrations before 1045 MST were 0.015 ppm at 0900 and 1000 MST. The maximum afterwards was 0.046 ppm at 1800 MST. While the SO_2 concentrations measured after 1045 MST were higher than the background levels, they were still considered to be low.

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* The plant elevation, not given in the report, is about 2100 m MSL.

All three surface SO_2 monitors were east or northeast of the plant; no surface samplers were located in the more mountainous terrain to the west of the plant where plume impingement on elevated terrain might be more likely to occur. It is not known whether any airborne sampling was conducted in the latter area; if it was, results were not discussed in the initial summary report. Owing to the limited nature of the surface sampling network, as well as the short time period characterizing the study as a whole, any conclusions which could be made from the data would be of limited significance with regard to the effect of complex terrain on the behavior of the Naughton plume.

FOUR CORNERS PLANT

Plume tracking studies covering somewhat similar time periods (less than one week) were conducted in the vicinity of the New Mexico Four Corners Power Plant by North American Weather Consultants (Niemann 1973) and Meteorology Research, Inc. (Smith and Anderson 1974). Both of the studies involved airborne sampling of pollutants (SO_2 in the first case and both SO_2 and NO_x in the latter case).

The objective of the MRI study was to test certain factors in the NOAA Model. These factors were: (1) The possible effects of buoyancy and exit velocity on the initial plume dilution, and (2) The subsequent rate of plume growth downwind. The objectives of the NAWC study were to measure: (1) the initial dilution factor in the initial momentum-dominated phase, (2) the plume centerline concentration, (3) plume geometry, (4) the crosswind concentration distribution downwind, and (5) the dissipation due to turbulence over flat and complex terrain.

Three days were selected as test cases by MRI: February 17, 18, and 19, 1974.* Terrain downwind of the Four Corners Plant for the February 19 case was not exceedingly rough, but the important features included a drop of 122 m in terrain within the first 1.6 km of the plant site, with several subsequent rises at various buttes within 16.1 km. Turbulence dissipation measurements were taken for each case by a "Universal Indicated Turbulence System" aboard the aircraft. Greater dilution rates were observed for February 17 and 18 than for February 19.

Considerable variation existed from one day to the next, depending on the particular roughness fetch, without any change in stability category as given by the temperature lapse rate. On February 17, concentrations remained lower than predicted throughout the entire observed plume travel (29 km), indicating enhanced dilution. Smith and Anderson considered turbulence measurements necessary in accurately estimating plume diffusion, particularly when the layer containing the plume is not connected to the surface layers by turbulent mechanisms (i.e., the plume was above the low-level drainage flow). They concluded that significant differences in plume dimension and dilution could occur without any major change in the stability category as a result of turbulence variations at the plume level.

Comparisons of normalized plume centerline concentrations of SO_2 (i.e., $\chi u/Q$ values) obtained aloft from airborne plume tracking observations under stable (E and F) conditions were made to those calculated using the NOAA model. For E stability, the model calculated values were greater than the observed values at a distance of 1 km downwind by a factor of about 5 on February 17, 1974, and by a factor of 15 on February 15, 1974 and February 19, 1974. For the F category, model calculated values for 1 km downwind were greater than observed by a factor of about 7 on February 17, 1974, and a factor of about 30 on February 15, 1974 and February 19, 1974. At distances approaching 12 km downwind the measured values approached the model predictions to a very close degree; within a factor of 2 for the E category and within a factor of 3 for the F category in all cases.

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* Data were collected on a total of five days - February 15, 17, 18, 19, and 20, 1974.

Niemann (1973) has compared instantaneous SO_2 plume centerline concentrations observed aloft for the Four Corners Plant under neutral and stable conditions to corresponding values calculated from the conventional Gaussian formula for flat terrain and from the stable version of the NOAA model, respectively. He found that both models overestimated the measured concentrations close to the plant by a factor of 10 (i.e., under both neutral and stable conditions), while at distances greater than 15 km the conventional Gaussian calculations generally agreed with observed values while the NOAA model overpredicted centerline concentrations by a factor of two under stable conditions.

These results agree at least qualitatively with the comparisons made by Smith and Anderson (1974), in that the Gaussian models show greater overestimation near the plant, while being within a factor of two at greater distances. In both works (by Niemann, and by Smith and Anderson) the authors attribute the initial overestimation to enhanced initial dilution from plume momentum and buoyancy. In one of the three stable cases examined by Smith and Anderson, and in the two cases examined by Niemann, the observed centerline concentrations at larger downwind distances remained less than the model values, by factors ranging from 2 to 5. Niemann suggested that this was a result of increased horizontal plume meander and oscillation, and vertical undulation over the complex terrain in the vicinity of the Four Corners Plant. Smith and Anderson have suggested that observed differences in turbulence dissipation rates accounted for the variations which they observed in downwind plume behavior under similar stability conditions (i.e., stable) as defined by the vertical temperature gradient.

In the February 19, 1974 case, near the stack (0.8 km), both the width and height of the plume exceeded the model values for both stability categories (i.e., σ_y by almost a factor of 3 for F stability). This effect was probably caused by rapid initial dilution of the plume by buoyancy and turbulent mechanisms which may or may not have been terrain induced. At 8.85 km downwind the observed lateral spread was still much greater than the model values (about a factor of 2), but the observed σ_z became less than either the model E or F values. The observed σ_y dimension was found to lie between the model E and F values at 29 km; the observed σ_z remained much less than the model value for either category (i.e., a factor of 3 less for E stability). Therefore, rapid growth of the plume occurred initially followed by much slower growth than predicted by the model for either E or F stability.

Assuming that σ_y and σ_z are power functions of downwind distance from the source, the following exponent powers were found to be applicable at downwind distances of 8.8 to 29 km compared to the powers assumed in the NOAA model for February 19, 1974:

For the relation:

$$\sigma(x) = x^p$$

x = downwind distance

	p = exponent	
	σ_y	σ_z
Observed	0.35	0.32
NOAA model E Stability	0.88	0.44
Model F Stability	0.88	0.38

These data indicate that the downwind lateral spread was much less than predicted by standard techniques and the vertical spread (σ_z) was somewhat less than suggested by the observed temperature lapse rate. In retrospect, had the additional time and financial resources been available, certain supplemental data would have been extremely helpful for further insights into the behavior of the plume. The NAWC study was carried out with limited meteorological observations, with one vertical temperature sounding being taken near the plant before each plume tracking flight and no upper level winds in the vicinity of the plant. Each of the

MRI flights consisted of more frequent temperature soundings at various distances downwind, and were supplemented by upper-level winds. With regard to the NAWC study, an additional downwind vertical temperature measurement would have been useful in at least one case, when the plume seemed to be fumigating to the surface after the lone early morning sounding had indicated a low level inversion. In neither study were sufficient wind data obtained to accurately determine flow characteristics over the entire area being sampled (e.g., the frequent upper level wind soundings taken in the MRI study were at one location, about 5 km north of the plant).

One of the two major objectives of the program of study conducted by Smith and Anderson (1974) at the Four Corners Power Plant was to investigate the dynamics of plume impingement on obstacles in the plume path. However, these investigators soon learned that only limited knowledge of the plume's impact on the surface could be gained without a ground-level sampling system. An important factor that affects the ground-level concentration is whether the plume remains in the drainage flow or is able to penetrate it into the synoptic flow above. Smith and Anderson concluded that for drainage flows of less than 1.5 m s^{-1} the plume would penetrate and rise above the drainage flow but absence of a ground sampling system prevented verification of this conclusion. The analysis of data was a bit more detailed in the MRI study; both observed horizontal and vertical plume dimensions (standard deviations) and normalized concentration data were compared to values predicted by standard Gaussian model values. However, the comparisons were only for a very limited range of meteorological conditions (stable lapse rate and light drainage winds). Although the NAWC study compared only observed and calculated centerline concentrations, the comparisons were for a greater (although not appreciably wide) range of meteorological conditions (neutral stability with modeeate winds and a stable lapse rate with light winds). Neither study encompassed more than a few days.

NAVAJO PLANT

The Navajo Plant, the largest electric generating plant in the State of Arizona, has been the subject of the greatest number of studies of plants operating in complex terrain. The research performed to date include daytime and nighttime tracer studies by North American Weather Consultants (NAWC 1974, Hovind et al. 1973b), extensive ground-level and aircraft monitoring of SO_2 (Rockwell International, MRI and SAI, 1975) and model-to-model comparisons of Navajo data (INTERA 1975).

The Navajo station is located at 1326 m MSL in a large natural basin with the Colorado River cutting across the center. Flat mesas rise up to 1850-2450 m MSL within 40 km of the plant, and Navajo Mountain (3166 m MSL) is located 48 km to the east. The two most significant terrain elements for plume impaction are Vermilion Cliffs 22.5 km to the west and Leche-e Rock 8 km to the southeast. The Vermilion Cliffs are part of a larger elevated landform (Paria Plateau) which rises somewhat abruptly in the vicinity of the Cliffs, while Leche-e Rock is a relatively narrow landform with a flat top; thus, these features represents two distinct types of elevated terrain which could conceivably alter the plume direction in different ways (i.e., the plume could flow around Leche-e Rock much more easily than around the Paria Plateau). The two locations also represent areas whose elevations are close to that of the predicted plume centerline under stable conditions.

The daytime tracer study (NAWC 1974) was designed for the purpose of evaluating the quantitative impact of fluorescent particles (FP), smoke, and equivalent SO_2 impact upon surrounding terrain as well as the documentation of possible unfavorable meteorological-topographic effects on plume rise, transport, and diffusion during conditions other than those postulated as 'worst case' (i.e., stable, light-wind) conditions in the NOAA (Van der Hoven 1972) Southwest Energy Study. The nighttime tracer study (Hovind et al. 1973b) had as its objective the simulation of plume transport and diffusion under postulated worst case (plume [illegible] conditions.

In both studies, FP tracer was released from the 236 m stack and from aircraft simulating an effective stack height of 210 to 250 meters above the stack. The period of the nighttime tracer program was characterized by nocturnal inversions extending above stack height, but not as high as the simulated effective stack height. Continuous recording of winds at stack top and numerous pibal and temperature soundings at various locations near the plant site were taken during the releases. The nighttime tracer study was supplemented by six continuously recording wind stations, most of which were located on elevated terrain, 8 to 40 km from the plant, which equaled or exceeded the effective stack height under stable conditions. Both the daytime and nighttime tracer studies were part of a larger diffusion climatology program conducted by NAWC at the Navajo site from May to September 1973. The meteorological data included pibal and temperature soundings, and surface and stack top wind measurements. The extensive meteorological data were used to construct time cross-sections of the wind flow fields for periods including the tracer releases. No attempt was made to quantify the tracer behavior in terms of model parameters which could use this data as input (i.e., plume geometry as a function of downwind distance, or various types of terrain). However, a Rotorod network strategically distributed around the Navajo Plant allowed for a quantitative evaluation of the impact of FP tracer upon the most prominent elevated terrain features at critical elevations between stack top and effective stack height.

The FP releases did not support the theory of direct centerline impingement on elevated terrain. In the night case of June 5-6, 1973, the maximum impact from a stack top release was on elevated terrain to the south and southwest of the plant within 24 km, but there was no distinct maximum impact zone. High FP counts were distributed over a large sector of elevations. On June 6-7, 1973, the FP plume was initially transported toward elevated terrain, but the plume never reached the surface. The authors have hypothesized that the plume may have become entrained in a downdraft associated with strong wind shear induced by the rising terrain, since the maximum impact was at an elevation lower than the release point. This phenomenon is evidence that strong vertical transport can occur near elevated terrain even in stable conditions. FP tracer releases showed greatest impact on the sampling network under light wind looping conditions on summer days. No quantitative information on the degree of turbulence enhancement by elevated terrain was given, however, the effect was expected to be less than for nighttime stable conditions.

The possibility exists, of course, that the plume centerline in the FP experiments may have reached the surface, but not directly over the network. Conditions in wintertime may be more stable and similar experiments may show even further enhancement of plume meandering. In the daytime study, plume impingement was investigated under limited mixing conditions and during inversion "break-up" conditions. Visual smoke plumes showed downwash within 1.6 km of the plant with strong winds. Smoke releases upwind of elevated terrain showed no direct plume centerline impingement as predicted by the NOAA model, as the plume was transported either over or around obstacles. The greatest impact on elevated terrain occurred with turbulent downward transport of the plume during transitional fumigation periods between stable morning conditions and neutral/unstable afternoon conditions. Plume meandering occurred frequently at Navajo under stable, light wind conditions due to interaction between the gradient flow and the diurnal thermal circulations induced by terrain.

Oil smoke was also released just upwind of Leche-e Rock and Kaiparowits Plateau, two prominent terrain features in the vicinity of the Navajo Plant. There was no evidence of direct centerline impingement on Leche-e Rock. However, later in the morning increased mixing associated with a transitional fumigation period allowed transport of the smoke into the rills of Leche-e Rock. At Kaiparowits Plateau, downslope drainage flow caught some of the smoke, but the remainder rose 305 m over the plateau and there was very little diffusion to the surface.

Hovind et al. (1973b) have made comparisons of NOAA model calculations with equivalent SO_2 ground concentrations determined from measurement of FP tracer concentrations resulting from releases from the Navajo stack (or by aircraft at the estimated effective plume height prior to the beginning of the plant's operation). The comparisons were for nighttime tracer releases under stable conditions, and for maximum hourly concentrations resulting from releases of 1 to 3 hours duration. For both the aerial and stack top releases, the NOAA model values ranged from a factor of about 20 to a factor close to 300 times greater than the measured equivalent SO_2 concentrations. The possibility of less than 100 percent dispersing efficiency was discussed in the NAWC report describing these comparisons; however, the point was made that even if an extreme value of 50 percent reduction in dispensing efficiency is assumed, the maximum SO_2 measured would still be nearly two orders of magnitude below the calculations. Their comparison between equivalent maximum hourly SO_2 concentration values and values predicted with the NOAA Impaction Model in 7 tracer releases show the model over-predicting by factors of 20 to close to 300.

INTERA Consultants (1975) have made comparisons between calculated SO_2 values, using the NOAA, EPA "VALLEY" and INTERA models, and maximum measured concentrations collected in the vicinity of the Navajo Plant. The SO_2 data representing Vermilion Cliffs were taken from five monitoring sites located in a line atop the Cliffs which are approximately crosswind when the wind is blowing from the plant toward the Paria Plateau. Hourly average SO_2 data from these five sites for the three days on which the highest SO_2 hourly averages occurred atop the Cliffs were compared with models. The highest hourly average concentration atop Leche-e Rock was also compared to model calculations. Aerial plume centerline measurements for each of these four days were additionally compared to values calculated by the standard Gaussian model (as presented in the EPA workbook, Turner 1970) and the INTERA model values.

The model input parameters for wind speed and direction were specified from pibal data near the plant site (Page Airport) and surface wind data (combined to estimate likely speed and direction at time of high concentration). Temperature soundings made mostly near the plant provided the basis for stability estimates. Comparisons are made for the range of stabilities and windspeeds thought to be appropriate for the travel time to Vermilion Cliffs. Limited mixing was assumed when appropriate, as in the Leche-e Rock case. The wind direction which provided the "best fit" of values calculated by the INTERA model to the observed data was used for all three models. The effective plume height was obtained from aerial SO_2 measurements on each day. The comparisons for the three Vermilion Cliffs cases and the Leche-e Rock case are shown in Table 12.

TABLE 12. MODEL AND MEASUREMENT COMPARISONS FOR PEAK HOURLY AVERAGE SO_2 CONCENTRATIONS (PPM) OBSERVED AT ELEVATED TERRAIN FEATURES NEAR NAVAJO GENERATING STATION

Site-Date	Stability (Windspeed, m/sec)	Model Calculations (ppm)			Maximum Measured Peak Hourly SO_2 (ppm)
		NOAA	EPA "VALLEY"	INTERA	
Vermillion Cliffs (10/16/74)	F (1.5)	1.25	0.20	0.11	0.102
	F (2.1)	0.89	0.14	0.08	0.102
	E (1.5)	0.46	0.11	0.078	0.102
	E (2.1)	0.33	0.08	0.058	0.102
Vermillion Cliffs (11/24/74)	F (1.5)	1.02	0.17	0.087	0.070
	F (2.1)	0.73	0.12		0.070
	E (1.5)	0.38	0.08		0.070
	E (2.1)	0.27	0.06	0.057	0.070
Vermillion Cliffs (12/2/74)	F (1.5)	0.80	0.13	0.066	0.069
	F (2.1)	0.57	0.09		0.069
	E (1.5)	0.31	0.07		0.069
	E (2.1)	0.22	0.05	0.048	0.069
Leche-e Rock (12/10/74)	Limited mixing (1.5)	0.39	0.21	0.16	0.071

Note: The EPA "VALLEY" model was not designed for the calculation of hourly averages, hence the results shown here for "VALLEY" are of limited relevance.

It is seen that for the input values specified, the NOAA model overpredicts by a factor of 3-15, while the VALLEY and INTERA models are generally within a factor of three. For the Vermilion Cliffs cases, the VALLEY model performs the best if E stability is assumed, while the INTERA model performs the best if F stability is assumed.

The comparisons with aerial measurements were made in each case for three or four downwind distances, generally between 3 and 22 km from the plant. The model calculations were expected to be somewhat lower than the measured values since the former corresponded to a one-hour average at a particular point and the latter to an "instantaneous" value. However, this was not the case for the aerial data collected on the high concentration days at Vermilion Cliffs. At closer distances to the plant (4-8 km), the INTERA model overpredicted the plume centerline concentration on all three days by a factor of 2 to 3 for the meteorological conditions under which it performs best with respect to ground level concentrations (F stability, 1.5 m/s); while approximately equaling the centerline values for E stability and 1.5 m/s. The INTERA model calculations for either condition at 5 km were generally within 20 percent of observed values but low by a factor of 2 to 4 at 22 km. The Gaussian calculations for either E or F stability (without sector averaging as employed by the EPA model) were somewhat higher than the INTERA model predictions (by up to a factor of 1.5) at close distances to the plant (about 8 km), while the calculations were of about equal value at greater distances.

Extensive ground-level and aircraft monitoring of SO_2 was undertaken near the Navajo Generating Station and the results were presented in a report assembled by Rockwell International, MRI, and SAI (1975). Since the purpose of the project was to determine if additional SO_2 removal would be necessary at the plant, field measurements were conducted during the season of the greatest occurrence of stable conditions (fall and winter), as determined from climatological records.

The principal measurements obtained from the field program included three-hour average stack emissions, ambient SO_2 concentrations, surface winds from a wide range of terrain elevations (including those potentially affected by plume impingement), pibal soundings at least four times daily (increased to 12 times daily during intensive study), and radiosonde and aircraft temperature soundings (once and twice daily, respectively). Particular to this study among all of those reviewed were the following:

- Classification by terrain location and meteorological conditions of the highest measured ground-level concentrations.
- Comparison of SO_2 peak (five minute) to mean (3 hour) ratios for elevated terrain near the Navajo Plant (high enough to be impacted by the plume) to equivalent ratios obtained by Montgomery et al. (1972) at TVA's Paradise Plant located in relatively flat terrain.
- Spatial interpolation of three-hourly SO_2 concentrations for samples located on elevated terrain (at or near plume impingement height) to estimate daily three-hour SO_2 maxima.
- Utilization of results from several previous field programs* conducted at the same site in the design of the field sampling program.

Vermilion Cliffs was the area most significantly affected, with direct impingement being the dominant mechanism. The study looked at three peak cases, and compared these high concentration days to 23 other non-zero low concentration days. It was concluded that the peak concentrations of SO_2, determined with respect to Q, resulted from: (1) the plume centerline being near the top of the cliffs; (2) a steady plume direction over at least a 3-hour period; and (3) light winds (1 to 4 m/s) at plume level. The major factor separating cases with slightly lower SO_2 values from the peak cases was variable wind direction. Cases with the greatest SO_2 impingement on the cliffs usually were characterized by the plume being forced down in front of the cliff.

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* Two of these studies were the daytime and nighttime tracer studies conducted by Hovind et al. which were previously discussed; the other two were background measurements obtained by Dames and Moore (1973), and another six-day field study conducted by Smith and Anderson (1975) which was not available at the time of this report but is summarized in the Rockwell report.

The prime requirement for significant SO_2 concentrations on Leche-e Rock was steady wind direction toward this area for several hours. This condition was met only twice during the program. Plume height and wind speed were then critical for determining the actual concentrations. Monitoring sites were located on top of each element of elevated terrain to insure interception of the plume if impingement occurred.

HUNTINGTON CANYON

Several independent tracer studies have been carried out in Huntington Canyon, Utah, a very steep-walled canyon with generally mountainous terrain.

A study by the Air Resources Laboratory of NOAA (Start et al. 1973) was conducted in March and April of 1973, with the purposes of (1) determining the nature and extent of plume impaction on elevated terrain and (2) comparing dilution within rough terrain to that expected over flat, open terrain. The instrumentation network was of appreciable density, consisting of (1) 40 SF_6 tracer gas surface samplers located at various points on the canyon walls and floor (sampling averaging time of about one hour), primarily up-canyon from the stack release point, and within 10 km of the latter; (2) nine surface wind speed and direction recording stations, six of which were up-canyon from the stack release point; (3) aspirated resistance thermometers at three levels on the 183 meter stack of a power plant which was under construction at the site; and (4) a Bell Helicopter which collected one minute plume samples. The number of SF_6 tracer releases was limited to 11, with 6 of the releases being from the elevated stack. The latter were made exclusively during daytime lapse conditions. One elevated release was made during inversion conditions, that being from a point of elevated terrain which jutted into the canyon.

Comparisons were made between observed centerline values and corresponding values calculated by the standard Gaussian plume model and stratified by stability categories.* Calculated versus observed plume center concentrations were found to range from minimal differences during moderate to strong temperature lapse, to canyon dilutions of five times that of "standard" flat terrain curves, to values about fifteen times more dilute than standard curves for strong inversions. The phenomenon of enhanced mechanical turbulence is believed to be responsible for the greater effluent dilutions with increasing stability in this study.

The data summarizing the results of the elevated release tests for D stability* are presented in Figures 14a and 14b from Start et al. (1973). Aerial centerline samples are represented by 0, ground-level samples are depicted by X; Figure 14a includes both helicopter and ground samples, while 14b represents helicopter samples only. Figures 15a and 15b are comparable to Figures 14a and 14b but are for ground-level releases for conditions of stable down-canyon flow. The solid curves represent the expected flat terrain concentrations.

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* See Table 11 for category definitions.

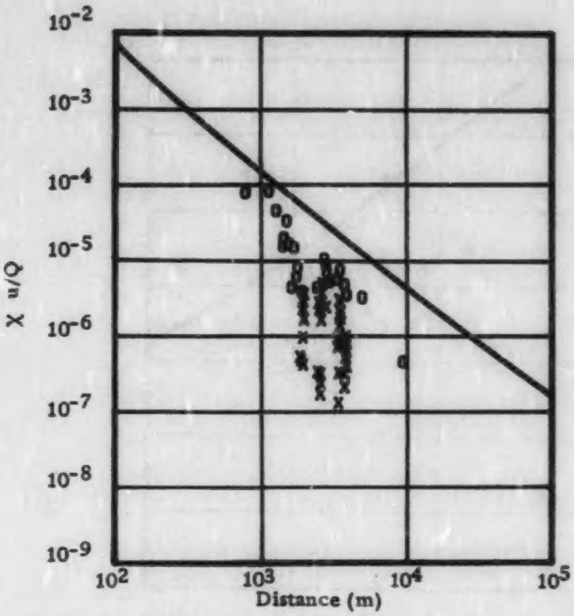

Figure 14a. Helicopter and Ground Samples for D stability.

0 - Aerial samples
X - Canyon wall and floor samples

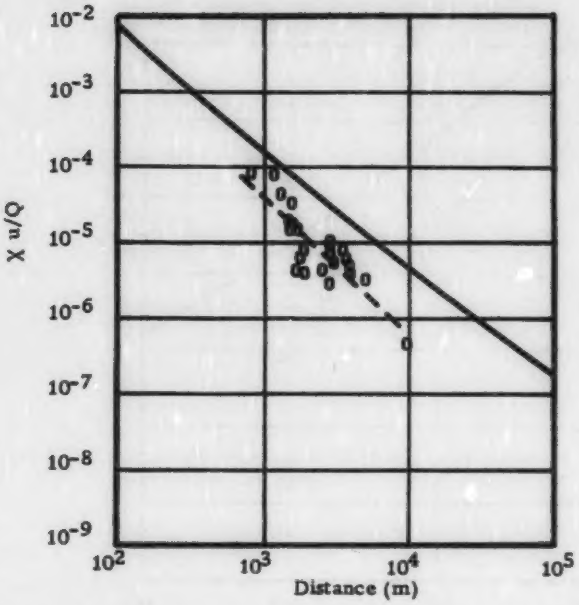

Figure 14b. Helicopter Samples only for D Stability Elevated Release Tests

0 - Aerial samples

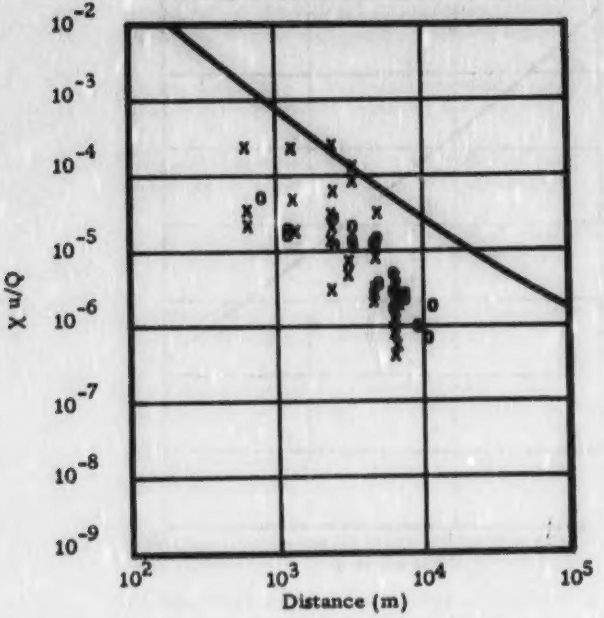

Figure 15a. Helicopter and Ground Samples for F Stability Release at Ground-Level in Conditions of Down-Canyon Flow

0 - Aerial Samples
X - Canyon Wall and Floor Samples

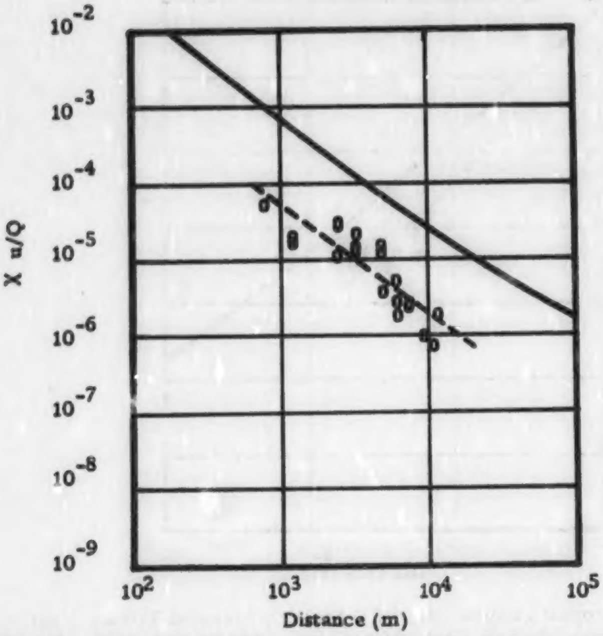

Figure 15b. Helicopter Samples only for F Stability Release at Ground-Level in Conditions of Down-Canyon Flow

0 - Aerial Samples

North American Weather Consultants (Hovind et al. 1973a) conducted a series of three separate tracer experiments, each of one week's duration, in order to provide a quantitative description of the air quality impact from a 430 MW power plant site in Huntington Canyon, and also to evaluate the applicability of the NOAA and Standard Gaussian plume centerline models.

Tracer Program III, the most extensive of the three tracer experiments, consisted of FP tracer and smoke releases from the estimated effective stack height as well as helicopter smoke releases near elevated terrain. The FP tracer concentrations were subsequently converted to equivalent SO_2 concentrations. Six surface stations monitored temperature and winds at various locations within the canyon, while pibal and aircraft temperature soundings were made up to three times a day. Included in the sampling network was a special sampling grid on one of the canyon walls aligned in a principally crosswind direction.

Aircraft measurements of the turbulence dissipation parameter (ε) were made with a Universal Indicated Turbulence System. This parameter, which is actually a measure of the turbulence dissipation rate in the inertial subrange of eddy sizes, indicates the rate of energy flow from larger to smaller eddies, and has been related to a turbulence magnitude scale. Values of ε were recorded that indicated moderate to heavy turbulence even when stabilities were classified as stable or extremely stable. Greater values of ε were found over Huntington Canyon than over Castle Valley, a nearby terrain feature characterized by less roughness.

In the plume tracer studies by Hovind et al. (1973a), the plume tended to follow a meandering path during up-canyon transport along a mean direction outlined by the elevated boundaries of the main and side canyon. Persistent plume transport in the stable air mass across Huntington Canyon toward the major topographic features was never observed, although the plume would at times move briefly toward that direction only to be deflected into an up-canyon direction while approaching the canyon walls. Therefore, the NOAA model assumption of plume transport across canyon onto elevated terrain was in this case unjustified.

During a week of observations in January 1973, all except one of the days were characterized by stable conditions, and the remaining day was neutral. Model-to-measurement comparisons by Hovind et al. (1973c) indicated that the NOAA model overestimated centerline concentrations by 10 times at 2.5 km downwind, but was closer to unity beyond 10 km from the plant site under stable, light wind conditions. No information was given regarding σ_y and σ_z, except that the plume influence on the sampling grid was rather uniform for stable conditions. This could either mean that σ_y was much greater than the lateral extent of the sampling grid (400 m) or that the plume was non-Gaussian. There is also serious question as to whether the aircraft release was equivalent to a point source, thus making comparisons to model calculations unjustified.

In another analysis effort by NAWC, Hovind et al. (1974) described studies of FP and oil fog dispersion that were undertaken at three sites with different roughness characteristics in the southwestern U.S. to determine the effect of terrain roughness upon plume dispersion from an elevated source. The three sites were characterized as follows: (1) a canyon with gradually rising canyon floors and steep walls; (2) a plant site located at relatively high elevation compared with surrounding terrain, with the general area being characterized by considerable ground roughness; and (3) relatively smooth terrain but with some low bluffs and cliffs in the direction of rising terrain. The authors do not disclose the names of the three sites in the report, although the first site seems representative of Huntington Canyon. Figure 16, from the report, provides a depiction of cross-section profiles at the three sites.

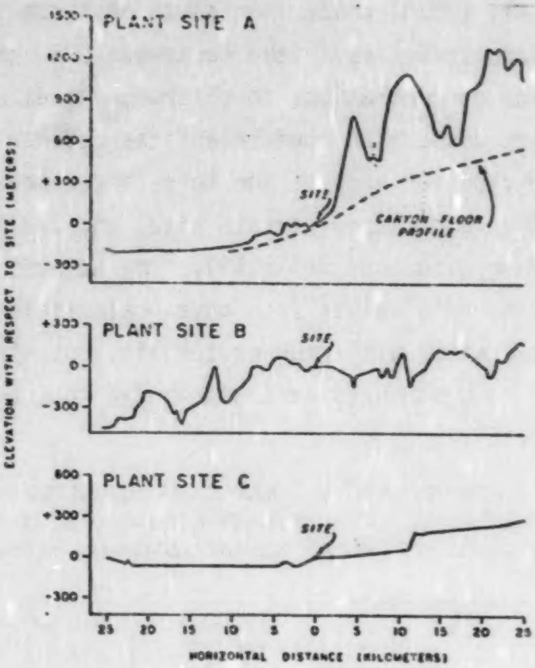

Figure 16. Terrain Cross-Sections at Three Sites of Varying Degrees of Roughness (from Hovind et al. 1974)

Aircraft FP tracer releases upwind of selected elevated terrain areas were employed at the first site. Six such releases were studied in January 1973, under stagnant atmospheric conditions with temperature inversions existing immediately above the release altitude. Oil fog was used as a tracer at the latter two sites. Data representing four cases characterized by neutral stability with moderate winds were summarized for the second site, while nine cases having a stable atmosphere were investigated for the site representing the smoothest terrain.

Hovind's et al. (1974) comparison study of three sites with different roughness characteristics in the southwest U.S. provided ratios of calculated centerline concentrations to observed values using a standard Gaussian model with dispersion coefficients as defined in Turner (1971). The average ratios for each of the three sites were 10.2 for the canyon site, 6.0 for the rugged open terrain site, and 1.2 for the relatively flat, open terrain site (see Table 13). The authors concluded that average departures of observed values from model calculations were significantly high for the two sites with rougher terrain, but minimal for the open, level terrain. The departures were attributed to increased turbulence in the rough terrain.

TABLE 13. AVERAGE RATIO AND RANGE OF CALCULATED TO OBSERVED RELATIVE CONCENTRATION VALUES χ' cal/χ'obs FOR THREE DIFFERENT TERRAIN CONFIGURATIONS (from Hovind et al. 1974)

	χ'_{cal}/χ'_{obs}	Pasquill Stability	No. of Cases
Confined Canyon (Site A)	$10.2\binom{7.4}{13.0}$	F	6
Rugged open terrain (Site B)	$6.0\ \binom{4.3}{7.5}$	D	4
Relatively flat, open terrain (Site C)	$1.2\ \binom{0.1}{2.4}$	E	9

For the canyon site the authors used σ_y and σ_z values corresponding to stability classes* C and D as inputs for the model, while observed meteorological conditions indicated an F stability condition. Since tracer releases were made below the inversion layer, the plumes, at least initially, were not in an atmosphere characterized by F stability.

Three mechanisms were hypothesized for the enhanced mechanical turbulence: (1) gradient flow momentum transport into the canyon; (2) drainage winds from feeder canyons which, when combined with additional slope drainage have enough momentum to be carried up the other side of the canyon; (3) turbulence effects created by the plume winding about protrusions or bluffs into the canyon center. However, there was no analysis of wind observations and no turbulence measurements to substantiate these flow hypotheses.

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* See Table 11 for stability class definitions.

The objectives of the study, to observe impaction and dilution within the canyon were met. It was concluded that transient impactions on the order of a few minutes may occur at any point during lapse conditions. The results are somewhat limited in significance because the topography of the release points varied greatly for the differing meteorological conditions and because sampling was only conducted for flow to the steep canyon terrain (the latter fact was acknowledged by the authors), i.e., no sampling was conducted to the northwest for flows which carried the plume toward the main valley to the east, away from the mountains.

Looking back upon the series of Huntington Canyon tracer programs conducted by NAWC, certain supplemental information gathering would have proven helpful with respect to evaluating the tracer impact under stable or very stable light wind conditions (e.g., those consistent with hypothesized worst case conditions). An extension of the study sampling period could have allowed for investigation of such phenomena as fumigation (observed in the NOAA study) or the possibility of lower level coupling with the gradient flow and subsequent cross-canyon flow. The aircraft FP tracer and turbulence measurements could also have been related in a systematic way to plume behavior (e.g., attempt to assess plume geometry from combined surface and aircraft FP data, and to relate plume centerline concentrations to turbulence measurements). An additional point to be made is that both the NOAA and NAWC Huntington Canyon programs involved tracer releases less than or equal to one hour in duration. Had these release time lengths been of longer duration, concentrations for longer averaging times might have been calculated, or peak to mean ratios (assuredly applicable for complex terrain) might have been utilized.

GARFIELD SMELTER

An investigation by NOAA ARL (Start et al. 1974) involved a short canyon near Garfield, Utah, in a mountain range along the south shore of the Great Salt Lake. The diurnal cycling of the lake-valley winds made this site more complex than Huntington Canyon. The tracer diffusion tests in this mountainous terrain involved the 122 meter Garfield Smelter stack and had the following objectives: (1) to measure ground-level and elevated centerline concentrations to quantify the plume dilution, if any, attributable to rough terrain; (2) to examine the degree of impaction against the mountain slopes; and (3) to establish a modest data base as an aid to mathematical analysis of diffusion in complex terrain.

Seven SF_6 tracer releases were made in as many days from the elevated stack. Forty samplers were positioned on the mountain slopes and canyon floor; in addition, 10 continuous wind speed and direction recorders were placed within the testing area, and pibals were taken both before and during the release period. Radiosonde ascents were also made while each test was in progress. The seven elevated releases were all made under similar estimated stability conditions* (B or C); two ground-level nighttime tracer releases (no visible tracer was released with the SF_6) were made under inversion conditions, but their discussion was limited in the report because of difficulties in locating a defined plume centerline.

Model-to-measurement comparisons of centerline concentrations and lateral plume dimensions were made for all of the elevated releases; for three of the releases, the σ_y values from the data were not derived from samplers positioned in a crosswind direction but rather from estimated plume widths or second moment** calculations. Similarly, σ_z values were derived from transformations of the Gaussian equation or with crosswind integrated concentrations; no actual measurements of vertical concentration gradients were made in the study. Plume measurements were not taken during early morning inversion conditions and so the effect of the drainage wind flowing out toward the lake could not be documented. Start et al. state in their report that "the plume impacted significantly" against the steep terrain on the west side of the canyon in one test case, while in other tests the plume conformed more or less to the terrain undulations, without significant impingement.

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* See Table 11 for stability definition.

** Second moment of the lateral distribution of tracer mass about the center of gravity of the mass distribution.

Based on visual observations during the tests, the authors suggested several mechanisms which are believed to contribute to the enhanced lateral spreading and lower centerline concentrations. These are summarized as follows:

- A plume approaching steeply rising terrain can experience lateral deflection in an attempt to flow around the blocking obstacle. This is particularly true during plume looping conditions when descending segments neared the ground.
- Mechanical turbulence, and vertical shear, believed to arise over and around the peaks and ridges of mountainous terrain results in an enhanced turbulent state (one visual observation of the actual plumes emitted from two of the smelter stacks revealed that the plume from the lower stack was being carried up-canyon while the plume from the higher stack was moving in a direction 90° from this, across a ridge in response to the near gradient flow aloft).

The results of Garfield aerial centerline measurements for stability classes B and C are exhibited in Figure 17a, b from Start et al. (1974). A least-squares, first-order curve fitting of the data points is represented by the dashed line in both graphs. Plume measurements conducted over level terrain are denoted by the O symbols in Figure 17b. The solid lines are the expected curves over flat terrain.

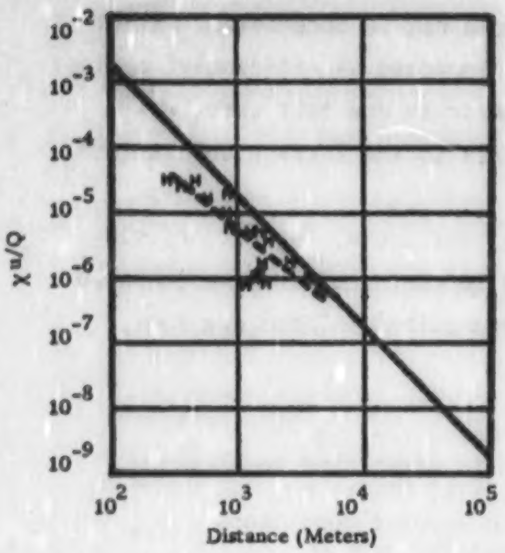

Figure 17a. Results of Garfield Aerial Sampling Test with Stability Class "B" Conditions

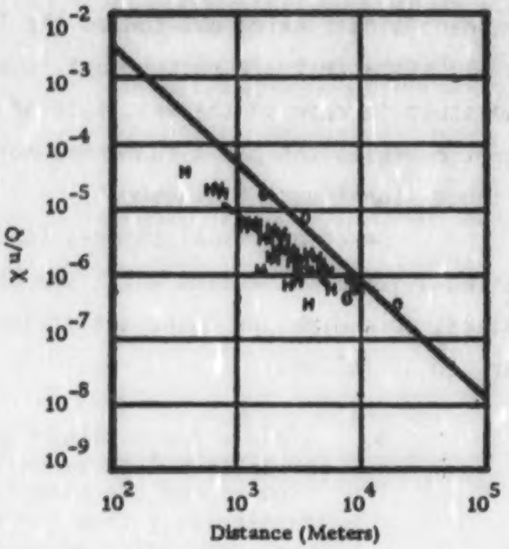

Figure 17b. Results of Garfield Aerial Sampling Test with Stability Class "C" Conditions

The researchers who conducted the Garfield SF_6 study (Start et al. 1974) made comparisons of observed normalized ($\chi u/Q$) plume centerline concentrations with calculations from standard curves presented in the EPA Workbook (Turner 1970). Observed and calculated lateral plume spreading were also compared. $\chi u/Q$ values of the seven elevated releases of SF_6 conducted in lapse conditions were two to four times less than predicted by the standard curves. However, data from four of the tests were not analyzed because of complicated plume geometry. In the three tests that did have a reasonably Gaussian shaped plume, σ_y exceeded the expected value by a factor of two. The enhanced lateral spreading may be due to avoidance of terrain by the plume. There was a tendency for the plumes to be deflected laterally in an attempt to flow out and around blocking obstacles. Values of σ_z however, were in general less than those for flat terrain. The aerial plume centerline comparisons were essentially the same as those described in the next paragraph, made by INTERA for the

NOAA model (since the latter is based on the same curves), and thus gave the same results. Lateral spreading was examined in a quantitative manner in the Start et al. (1974) study by (1) selecting slices across the isopleth patterns which were normal to the observed direction of plume travel; (2) extracting analyzed concentration values along each slice to obtain reasonable approximations of the concentration distributions; and (3) calculating σ_y values by both the plume "width" method and the second moment method (both methods resulted in σ_y values which were approximately the same). A comparison of these observed values with standard σ_y values for the appropriate stability classes (B or C) showed the average ratio of observed σ_y to Pasquill-Gifford σ_y to be 1.79.

The data from these three NOAA Garfield tests were also compared with the NOAA, EPA "VALLEY," and INTERA model calculations (Intera Consultants 1975).

The input data were determined as follows:

- The plume rise was obtained by extrapolating the plume centerline as determined from helicopter observations, back to the source.
- The INTERA model calculations were performed using wind profiles (1) from pibal data at the site and (2) from a standard power law for unstable conditions.
- The stability class was determined from the objective criteria suggested by Turner* (1970).

The models were compared by INTERA Consultants (1975) by using two different statistical measures. One method, involving the ratio of the calculated mean value to the observed mean value ground-level concentration, tends to give a greater weight to the larger measured and calculated values. This ratio (R1) can be mathematically stated as:

$$R1 = \frac{\Sigma X_{CALC}}{\Sigma X_{OBS}}$$

---

* See Table 11 for criteria used.

where

χ_{CALC} = calculated concentration
χ_{OBS} = observed concentration.

The second method gives an average of the calculated to observed ratio at every specific sample point. This logarithmic mean ratio (R2) is mathematically described as:

$$R2 = \exp\left[\frac{\Sigma \ln (\chi_{CALC}/\chi_{OBS})}{N}\right]$$

N = number of points.

The log mean ratio gives approximately equal weight to large and small measured and calculated values. Model-to-measurement comparisons for ground-level concentrations are summarized in Table 14.

TABLE 14. GARFIELD DATA MODEL-TO-MEASUREMENT COMPARISONS OF GROUND-LEVEL CONCENTRATIONS

Test	Estimated Pasquill Stability	Model	R1	R2
2	C	INTERA - Pibal	1.66	1.13
		INTERA - Standard	0.92	0.82
		NOAA	0.26	0.20
		EPA VALLEY	0.36	0.36
3	C	INTERA - Pibal	0.57	0.40
		INTERA - Standard	0.45	0.44
		NOAA	0.35	0.26
		EPA VALLEY	0.34	0.36
7	B	INTERA - Pibal	2.50	1.56
		INTERA - Standard	1.50	1.25
		NOAA	1.50	0.78
		EPA VALLEY	2.67	2.23

Analysis of these three tests, using R1, show the INTERA model ground concentration results to be within a factor of 2.5 and the results of the NOAA model to be within a factor of 3.9. For Test 7, the NOAA model performs as well as the INTERA model when the standard profile was input to the latter (R1 = 1.50), and better than the INTERA model with a pibal wind profile.

Additional comparisons were made with observed data in the three tests for predicted plume centerline concentrations as a function of downwind distance, and for how well the observed concentration patterns were reproduced spatially.

The helicopter samples, corresponding to an averaging time of the order of one minute, were converted to one hour values by the EPA Workbook technique (Turner 1970) to correspond to the model calculations, which represent one hour averages. The results of the comparisons for the aerial centerline data are as follows:

- The INTERA model calculations using standard wind profile data were lower than observed values by factors from 1.3 to 4.0 in two of the cases and were within a factor of 1.5 in the last test.
- The INTERA model values calculated using pibal wind data were within a factor of 1.5 in two of the three tests, but were higher than plume centerline values by a factor of 2 to 4 at distances beyond 1 km in the last test.
- The NOAA model calculations were greater than observed centerline values by a factor of 2 to 5 at distances within 4 km of the plant in two of the three tests and for all distances in the last test; calculated values approached observed values at distances greater than 4 km in Tests 2 and 3.

The results of the model comparisons for accuracy of reproduction of the observed concentration pattern showed the INTERA model predicting the location of the maximum concentrations accurately in two of the three tests, while the NOAA model was accurate in this regard in only one of the tests. The NOAA model predicted the location of the maximum concentration in a valley in two of the three cases.

Comparison of the heights of the maximum in-plumes concentrations indicate that the NOAA model assumption of parallel to terrain plume centerline flow in complex terrain under unstable conditions is invalid for the unstable conditions characterizing the days evaluated. For these days, the heights of the maximum in-plume concentrations over the ridges was in general half those over the valleys. The INTERA model usually predicted the heights with good accuracy, although it did not show quite as much conformance to the terrain features as actually observed.

KAIPAROWITS

A recent FP and oil fog tracer study has been conducted by North American Weather Consultants (1974) in the vicinity of the proposed Kaiparowits Generating Station which was to be located in the rugged canyonland of southern Utah. Some of the details of this study were not available at the time of this report. However, it is known that the study was fairly limited in duration, consisting of 10 FP tracer releases in May of 1974, 7 at one location in the area and 3 at another. Each of the releases was supplemented by measurements of the vertical temperature profile, vertical wind profile, and turbulence measurements. FP recoveries from four elevated tracer releases made by aircraft, two of which were at one proposed site (Fourmile Bench), and two of which were at the Nipple Bench site, were chosen for evaluation. One of the Fourmile Bench tests was characterized by early morning stable conditions, while the other three tests were characterized by unstable conditons. The principal interest in the study lies in the subsequent use of the data in model-to-measurement comparisons by Hoffnagle et al. (1975).

Initial model calculations were made using the stability category observed at the time of each test, and pibal wind profiles constructed for each test. Once the initial model calculations had been compared to the measured data, the models were adjusted individually to provide the best possible simulation within the limits of model flexibility.

Hoffnagle et al. (1975) have tested the EPA "VALLEY," NOAA, TVA coning model and INTERA models against fluorescent particle tracer data obtained by North American Weather Consultants at two proposed sites for the Kaiparowits Generating Station. It is important to note that the TVA model (which, like the NOAA model, is actually a set of Gaussian calculational techniques, with each technique being appropriate to a different meteorological condition) was not specifically designed for application in complex terrain situations; however, the comparison here is of interest because the model differs from the other standard Gaussian models (EPA "VALLEY," NOAA) in that the diffusion coefficients used (σ_y and σ_z) are not the same as the standard coefficients as described in Turner (1969), and were essentially derived from observations of power plant plumes. A different set of equations is used for computing maximum SO_2 concentrations from power plants under coning (neutral stability), inversion breakup, and limited mixing conditions; the INTERA comparisons were based on TVA "coning" model calculations in each case.

Initial model calculations were made using the stability category, observed at the time of each test, and pibal wind profiles made during each test (for determination of wind speed in the case of the Gaussian models, and wind profile in the case of the INTERA model). Once the initial model calculations had been compared to the measured data, the models were adjusted individually to provide the best possible simulation within the limits of model flexibility.

Table 15 shows the results of the comparison of initial model calculations with the FP data.

TABLE 18. COMPARATIVE MODEL RESULTS OF THE RATIOS OF THE MEAN CALCULATED TO MEAN OBSERVED VALUES OF EACH OF FOUR TRACER TESTS NEAR KAIPAROWITS (after Hoffnagle et al. 1975)

Model	Tracer Test #1		Tracer Test #2		Tracer Test #3		Tracer Test #4	
	No. of Receptors Evaluated	Ratio	No. of Receptors Evaluated	Ratio	No. of Receptors Evaluated	Ratio	No. of Receptors Evaluated	Ratio
NOAA	20	0.7	12	1.3	18	0.8	20	1.2
TVA	20	0.5	12	0.4	18	1.5	20	0.3
VALLEY	8	1.5	3	2.3	7	1.1	6	0.8
INTERA	20	1.7	12	1.7	18	1.9	20	0.9

- Of the 3 to 20 receptors evaluated in each case, the NOAA model had the best mean ratio of calculated to observed concentrations in three of the four cases, and its mean ratio was within 30 percent of unity in all four cases.
- None of the remaining three models had consistently better mean ratios than the other two. The TVA coning model generally underestimated (three of the four cases); the EPA "VALLEY" model had mean ratios from 0.8 to 2.3; and the INTERA model had mean ratios from 0.9 to 1.9.
- None of the four models had consistently good correlation coefficients, and thus did not accurately calculate plume behavior as a whole. The INTERA model had the highest single case correlation of 0.8. The INTERA, TVA and NOAA models had correlations exceeding 0.5 in only one case, while the "VALLEY" model always gave correlations below 0.5. (But it should be noted that the "VALLEY" model was never intended for use in short-term predictions.)

The "plume" recoveries in at least one of the cases (the release under assumed stable conditions) suggest that inversion breakup may have been occurring; the fact that no meteorological data were available to confirm this (as had been the case in the comparisons for the Navajo Generating Station) points out the need for better documentation. It also suggests that application of TVA's "inversion breakup" Gaussian equation might have been more appropriate in this case. The results of the other cases imply that the terrain in the Kaiparowits area is characterized by more enhanced dispersion for the given situation than would be expected from conventional Pasquill-Gifford measures of stability.

LAPPES PROGRAM

Description of the Site

The Large Power Plant Effluent Study (LAPPES) was initiated during 1967, and conducted during a subsequent five-year period, to evaluate the extent and effects of air pollution resulting from three coal burning generating stations with tall stacks (Conemaugh, Keystone, and Homer City) in rolling to mountainous terrain in Western Pennsylvania. The four-volume report on the LAPPES program by Schiermeier et al. (1970-1972) dealt mainly with data presentation and was not concerned for the most part with any data analysis, other than to qualitatively describe a few obvious cases of terrain influence on dispersion from the Conemaugh Station (located in the roughest terrain of any of the plants) as evidenced from airborne monitoring near that plant during April and October of 1971. The terrain in the vicinity of the Keystone, Homer City and Conemaugh Plants is composed of rolling hills rising 100 - 200 meters above valley floors and ridges rising 200 - 450 meters above valley floors. The major features are Chestnut Ridge, oriented NE-SW between the Homer City and Conemaugh stations, and Laurel Ridge which is considerably higher and located to the SE of Conemaugh.

The Keystone Station is situated in a shallow rural valley with two 244 m stacks the base of which are at 305 m MSL. Except for this valley, the surrounding terrain within 5 km is hilly, with the highest peak reaching no more than mid-stack height, i.e., about 120 m above the plant. The Keystone Power Station is about 25 km to the northwest of the closest ridge (Chestnut Ridge). So the influence of topography was not evident from ground-level measurements near Keystone Plant.

The Homer City Station, with twin stacks at a height of 244 m, is located on a plateau, with the surrounding terrain falling off as much as 100 m from the stack base height of 366 m MSL. The highest peaks within 5 km are those of Chestnut Ridge to the SE, which range slightly higher than midstack height, i.e., about 170 m above the plant. East of the ridge is another plateau which received higher SO_2 concentrations at ground level than were found at a similar distance in any other direction. This phenomenon may have been a lee effect of Chestnut Ridge.

The Conemaugh Station is the most susceptible to terrain influences. Separating this plant from Johnstown to the SE is Laurel Ridge, which has peaks reaching 180 meters above the stack top of 305 meters within 10 km of the plant. These hills along with the slightly lower Chestnut Ridge to the west may cause channeling of the plume. Schiermeier (1972) observed that under neutral conditions flow from the SE quadrant (passing over Laurel Ridge before reaching the plant) brought the plume to the surface very quickly. Ground-level concentrations then diminished with further distance, but then increased again on the lea side of Chestnut Ridge. Downwash on the lee side of Laurel Ridge, confirmed by subsidence of pilot balloons in the vicinity of the plant, was responsible for bringing the plume to the surface near the plant. Surface heating on sunny days precluded downwash. However, under northwest flow the plume rose over Laurel Ridge and mixed in a deep layer in the lee of the ridge. Only low concentrations were measured at ground level. No lee waves were detected downwind of Laurel Ridge. This lack of downwash supports the idea that roughness fetch plays a large role with regard to plume transport and dispersion characteristics. It is known from previous studies (Egan 1975) that flow separation in complex terrain may either take the form of large, reverse, stationary eddies, or "shed" vortices. It is conceivable that the former was being observed near the Conemaugh Plant with southeast winds (the downwash conditions), while upwind roughness characteristics favored the second type of separation flow with northwest winds, since downwash was then absent.

Terrain Roughness Parameters

In the course of preparing this present report GEOMET undertook special analyses of data from the LAPPES and other studies to assess the impact of terrain-induced turbulence on the diffusion parameters σ_y and σ_z in a Gaussian dispersion model. In order to quantitatively measure the terrain variation, ground elevations beneath several plume centerline paths were plotted from USGS topographic maps. The standard deviations of the elevations along the plume centerline paths out to various distances were computed. A space interval of 200 m was used for plotting elevations. Calculated Standard Deviation of Elevations (SDE's) are tabulated for comparison (Table 17) with the differences between plume parameters measured in the plume and conventional values for the same plume estimated by the methods of Turner's Workbook (1970).

Figure 18 shows the terrain cross-sections of 14 plume paths, plotted in various compass directions from the Keystone plant out to 16 km distance, together with the corresponding SDE values. The abscissa shows downwind distance in kilometers; the ordinate shows elevation above sea level in meters. (Note that the vertical to horizontal scale ratio is 0.06). The figure shows that larger SDE numbers correspond to more complex terrain. For example, the highest calculated SDE value, 58.2 m, is noted for the direction toward the east; the smallest calculated SDE value, 24.3 m, occurs to the south-southwest. The elevation profile to the south-southwest contains smaller variations than that of the profile toward the east. Figure 18 is included as an example of the technique for characterizing terrain cross-sections. Similar calculations were made for the other SDE values shown in Tables 16 and 17.

Plume Cross-Sections and Meteorological Data

Standard deviations (σ_y and σ_z) of the observed SO_2 concentrations in LAPPES plume cross-sections were statistically computed by GEOMET through integration of the vertical concentrations and crosswind concentrations

TABLE 16. DISPERSION PARAMETERS σ_y AND σ_z DERIVED FROM PLUME CROSS-SECTION MEASUREMENTS IN THE LAPPES PROGRAM

Plant	Wind Sector*	Distance From Plant (km)	SDE (m)	Field Test	σ_y (m)	σ_z (m)
Keystone	1	4	27.7	1	325.0	49.2
				2	513.3	60.3
				3	570.9	90.9
				4	289.1	44.1
				5	269.7	33.6
		10	33.4	1	867.9	41.5
				2	663.9	44.7
				3	1336.7	43.7
				4	1380.9	65.8
				5	1152.1	52.1
		16	33.7	1	2065.0	74.5
				2	1731.5	67.8
				3	1034.0	42.0
				4	384.5	20.0
				5	1578.1	57.6
				6	931.8	33.8
				7	765.7	75.7
	2	4	16.8	1	192.3	46.2
				2	427.1	87.1
		10	16.8	1	886.8	58.2
				2	434.9	58.3
				3	851.3	65.4
				4	767.2	112.9
				5	1206.9	49.0
				6	1268.7	57.8
		16	23.1	1	1217.9	118.0
	3	4	30.5	1	636.3	102.4
				2	902.4	46.7
		10	37.3	1	385.6	61.9
				2	532.6	73.5
				3	954.5	36.6
		16	37.3	1	603.7	34.9
				2	1024.4	1114.6
				3	553.4	160.9
				4	1216.1	51.2

(Continued)

* Wind sectors are measured from the power plant according to the 16 compass directions.

TABLE 16. (Continued)

Plant	Wind Sector	Distance From Plant (km)	SDE (m)	Field Test	σ_y (m)	σ_z (m)
Keystone	4	16	37.9	1	417.8	50.6
	5	4	25.2	1	473.9	41.3
				2	291.5	103.6
				3	365.9	64.7
				4	386.6	80.4
		10	33.3	1	687.5	60.2
				2	527.0	57.4
				3	561.9	108.4
		16	58.2	1	783.1	52.2
				2	757.5	34.9
	6	4	32.4	1	276.3	53.6
	7	10	40.0	1	441.5	58.7
		16	36.5	1	1807.3	78.2
	8	4	27.1	1	565.3	63.3
	10	4	19.8	1	881.7	29.4
				2	220.0	49.2
		10	24.3	1	1699.9	66.2
	11	10	31.5	1	378.0	48.5
		16	30.6	1	558.8	55.9
	13	4	34.6	1	220.9	109.4
		10	32.3	1	546.1	47.4
	14	4	42.0	1	261.4	88.7
				2	672.5	84.4
		16	42.5	1	553.5	63.7
	15	4	33.1	1	406.4	121.3
				2	634.4	108.6

(Continued)

TABLE 16. (Concluded)

Plant	Wind Sector	Distance From Plant (km)	SDE (m)	Field Test	σ_y (m)	σ_z (m)
Keystone	15	10	31.0	1	880.4	81.4
		16	30.1	1	841.1	78.6
	16	10	33.7	1	336.2	39.6
Conemaugh	1	10	66.6	1	781.6	46.8
	2	4	43.1	1	322.5	45.2
	3	4	40.8	1	304.8	38.8
				2	321.4	42.8
				3	280.7	62.6
		10	57.9	1	1109.1	79.8
	4	4	25.4	1	357.4	48.9
				2	515.9	73.2
				3	386.5	56.6
				4	314.4	74.0
		10	92.3	1	657.1	39.4
	5	10	175.3	1	1169.4	90.5
	6	4	105.0	1	465.1	93.9
	7	4	117.6	1	587.8	91.3
		10	166.8	1	1142.3	74.1
	15	16	65.0	1	1461.2	106.2
	16	10	66.8	1	945.1	79.3

TABLE 17. SUMMARY OF OBSERVED AND STANDARD σ VALUE, OBSERVED MINUS STANDARD, AND EQUIVALENT PASQUILL STABILITY CLASS FOR COMBINED KEYSTONE AND CONEMAUGH DATA

Travel Distance (km)	Number of Cases	SDE (M)	σ_y (m)				σ_z (m)			
			Observed	WADE Estimate*	Observed Minus Estimate	KY**	Observed	WADE Estimate*	Observed Minus Estimate	KZ**
4	4	16.8- 19.8	430.3	239.3	191.0	2.6	53.0	77.5	-24.5	4.9
	14	25.2- 27.2	401.8	239.3	162.5	2.8	63.1	77.5	-14.4	4.4
	6	30.5- 34.7	512.8	239.3	273.5	2.1	90.4	77.5	12.9	3.8
	5	40.8- 43.1	371.7	239.3	132.4	3.0	86.6	77.5	9.1	3.9
	2	105.0-117.6	526.4	239.3	287.1	2.0	92.6	77.5	15.1	3.8
10	7	23.1- 24.3	1016.5	543.6	472.9	2.4	66.8	134.9	-68.1	5.3
	12	31.0- 33.7	776.5	543.6	232.9	3.2	57.6	134.9	-77.3	5.6
	4	37.3- 40.0	578.5	543.6	34.9	3.8	57.6	134.9	-77.3	5.6
	3	58.2- 66.8	945.3	543.6	401.7	2.6	68.6	134.9	-66.3	5.2
	1	92.3	675.1	543.6	131.5	3.5	39.4	134.9	-95.5	6.0
	2	166.8-175.3	1155.8	543.6	612.2	2.1	82.3	134.9	-52.6	4.9
16	10	28.5- 30.7	1110.8	825.1	285.7	3.3	62.4	176.0	-113.6	5.8
	7	36.5- 42.5	882.3	825.1	57.2	3.9	79.2	176.0	-97.8	5.3
	3	58.2- 65.0	985.7	825.1	160.6	3.5	43.6	176.0	-132.4	6.0

* Estimate from Workbook for Atmospheric Dispersion Estimates (Turner 1970).

** KY and KZ are the approximate Pasquill Stability Class (i.e., A = 1, B = 2, etc.) required to represent the observed value.

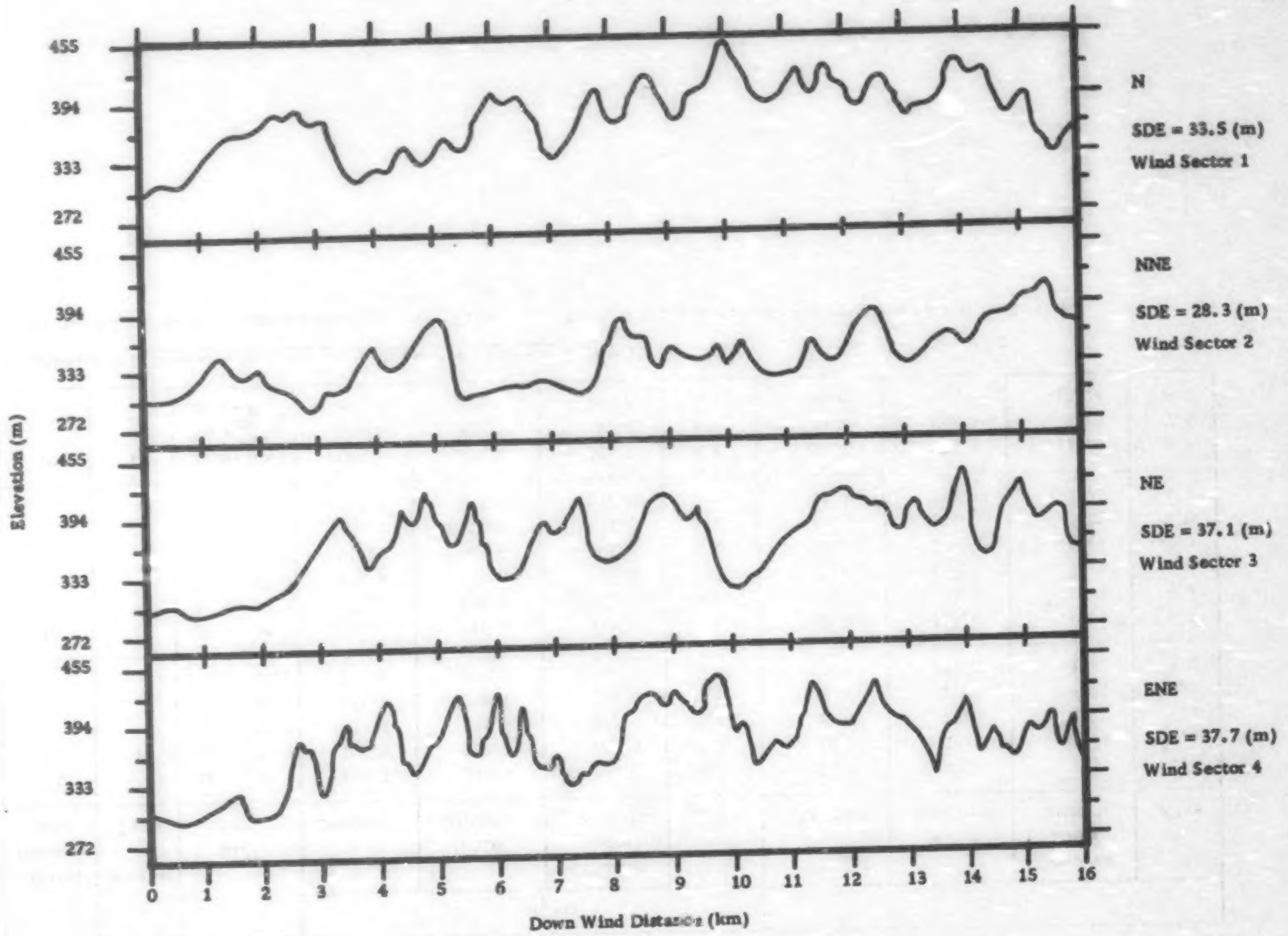

Figure 18 . Profiles of Elevation Toward 14 Directions from Keystone Power Plant. Standard deviation of elevation read by an increment of 200 m horizontally are also indicated. (Continued)

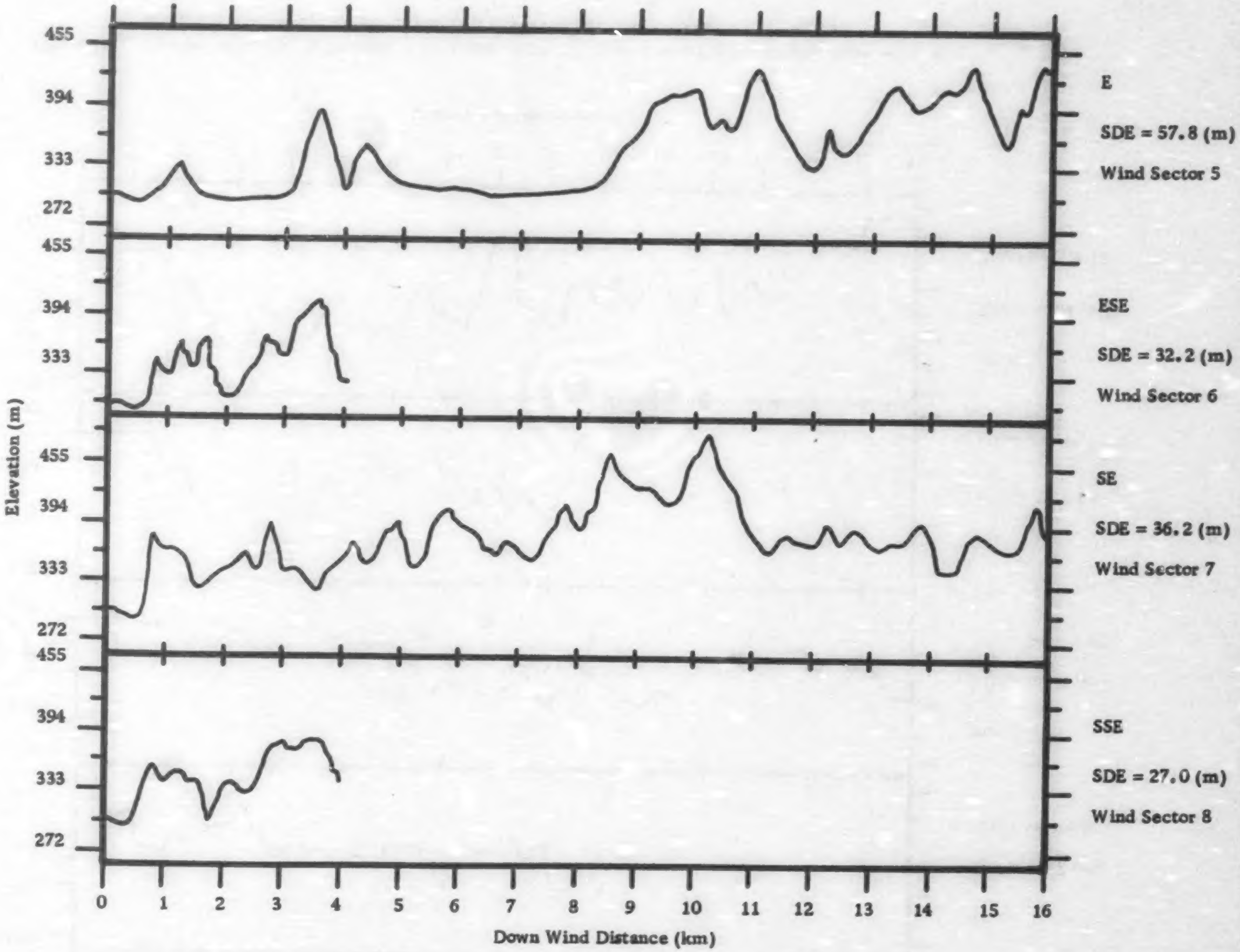

Figure 18 . Profiles of Elevation Toward 14 Directions from Keystone Power Plant. Standard deviation of elevation read by an increment of 200 m horizontally are also indicated. (Continued)

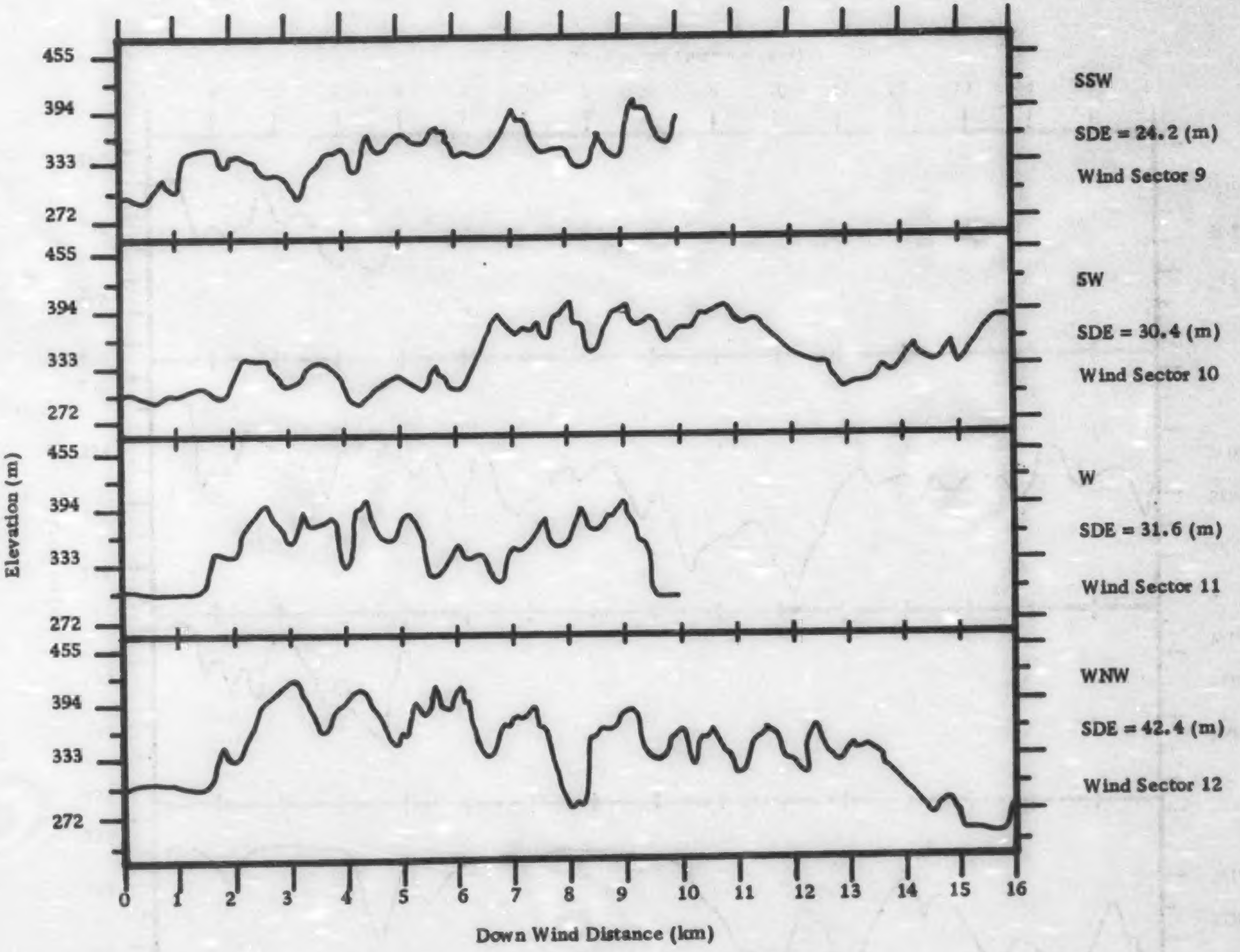

Figure 18 . Profiles of Elevation Toward 14 Directions from Keystone Power Plant. Standard deviation of elevation read by increment of 200 m horizontally are also indicated. (Continued)

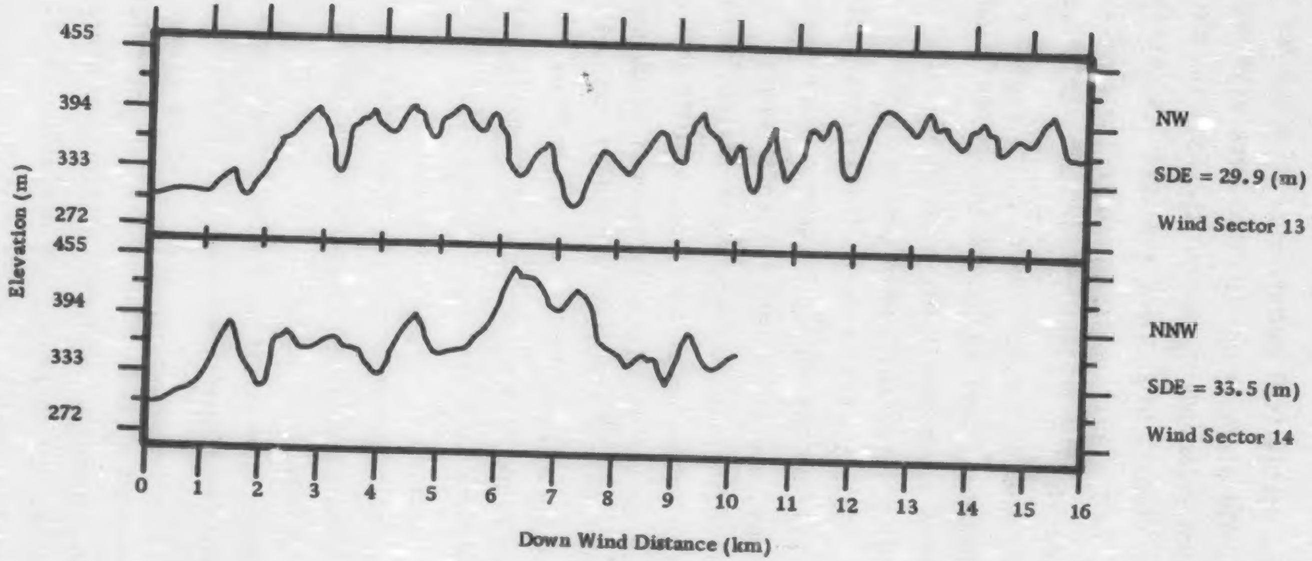

Figure 18 . Profiles of Elevation Toward 14 Directions from Keystone Power Plant. Standard deviation of elevation read by an increment of 200 m horizontally are also indicated. (Concluded) respectively. Because the statistical method used is valid for Gaussian distribution curves only, 436 plume cross-sections were examined manually and 28 percent (122), which appeared to be near the Gaussian bell shape, were selected for further study. Wind speed and solar radiation, measured at Jimmy Stewart Airport in Indiana, Pennsylvania in the LAPPES program, were used to determine the stability classes* for each measurement period.

Skewness was calculated for horizontally integrated concentrations measured at the Conemaugh Power Plant. Although the plume cross-sections used were selected to be nearly Gaussian in shape, vertical concentrations (horizontally integrated) were generally skewed toward the upper half of the plume. Seventy-two percent of the cross-sections were skewed toward the upper half of the plume, 17 percent were skewed toward the lower half of the plume, and 11 percent had no skewness. It is our speculation that these results indicate more rapid mixing in the lower portions of the plume than in the upper portions, perhaps due to a terrain-induced turbulence effect or due to thermally induced turbulence due to heating of the ground surface. A stronger possibility may be that the skewness was caused by a shearing-off of plume remnants as the main part of the plume rose to its buoyant level or into an inversion aloft.

Dispersion parameters, σ_y and σ_z, in a Gaussian dispersion model are considered to be a function of downwind distance and atmospheric stability. Conventionally, atmospheric stability is treated as a function of wind speed and solar radiation or lapse rate. This primarily represents the effects of thermally-induced turbulence; but the terrain-induced turbulence, which is very important in a complex terrain area, is not accommodated by the Gaussian dispersion model. If terrain-induced turbulence has no effect on plume dispersion, then we would expect that the differences between observed σ_y and σ_z and the values calculated by conventional methods (i.e., Turner's Workbook) will on the average be zero. If significant differences are observed, the deviation should correlate with terrain characteristics such as the SDE values.

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* See Table 11 for category definitions.

Plume measurements were sorted by their centerline direction into 16 compass directions. Observed values of σ_y and σ_z were averaged for each stability class, plume travel direction and distance. All cases selected were for neutral conditions (class D). Homer City data were omitted because of the small number of observations. Table 16 shows observed σ_y and σ_z values and SDE values for each plume-centerline direction and distance from the plant. In order to see the influence of SDE on σ_y and σ_z, we grouped the data by ranges of SDE values and downwind distances (4, 10 and 16 km) and computed the mean σ_y and σ_z for each group in Table 17. Also tabulated are σ_y and σ_z values which have been measured, values estimated from Turner's Workbook (1970), and their differences. The measured σ_y and σ_z values were used to determine equivalent stability classes, KY and KZ, based upon the observed horizontal and vertical concentration distributions, respectively. These values are also tabulated in Table 17. From this table one can observe that the impact of terrain on the horizontal dispersion parameter (σ_y) is more significant than on the vertical dispersion parameter (σ_z). If one examines the average of KY values for a fixed travel distance, an increase in the average KY value with distance is noted. At 4 km distance, the average KY is 2.5, while out to 16 km distance, the average KY is 3.6; this is closer to the actual stability class of 4. This indicates that the terrain influences on σ_y are more significant at closer distances from the plant; the terrain influence on σ_z is more important at further distances from the plant. With increasing distance the influence of terrain on the plume becomes smaller. The correlation between SDE and KZ is not recognizable. It should be recognized that these calculated SDE's depend to some extent on downwind distance and Δx (the distance increment) as well as the nature of the terrain; instead of a specific direction, consideration of a sector may be warranted.

KINGSTON POWER PLANT

The TVA's Kingston Power Plant has been the focal point for several intensive field programs. These programs exceed the magnitude of the Navajo and LAPPES studies in scope of data collections. A computer analysis of Kingston Plant data similar to the analysis of LAPPES data was performed by GEOMET to examine terrain-influenced dispersion within the plume; this work is described below.

Description of the Site and Monitoring Program

The Kingston Steam Plant is situated in eastern Tennessee about 56 km west-southwest of Knoxville and 3.2 km northeast of Kingston. The plant lies on a peninsula formed by the junction of the Emory and Clinch Rivers. Flowing southward from the plant site, the Clinch River combines with the Tennessee River some 4.8 km downstream. Within a radius of 1.6 to 4.8 km, the plant is almost totally encompassed by ridges 107 to 122 m higher than the plant grade. The terrain continues to be hilly outside the immediate area, with ridges aligned northeast-southwest and extending to elevations about 244 m higher than the plant grade.

Intensive field studies of plumes were conducted in the vicinity of TVA's Kingston Power Plant for three two-to-three month periods in the winter and early spring of 1973 - 1975. Only preliminary analyses of data had been made at the time of this report and these were essentially limited to calculations of σ_y. The most impressive aspect of this field monitoring program compared to the others previously described was the number of different modes of SO_2 monitoring used, and more importantly, the coordination between these modes which resulted in use of each to its optimal advantage.

For example, in the 1975 sampling period, a fixed-wing aircraft, van, helicopter, and 11 fixed surface monitors were used on a daily basis, in addition to plume photography. The van and helicopter were both equipped with SO_2 monitors and served to locate the portable monitoring in areas of high SO_2 concentrations. Airborne crosswind plume measurements were made at fixed distances downwind. Thus, the analyses of plume geometry could be made for this two-month period in late winter-early spring. The three data collection periods featured intense collection (from the standpoint of temporal resolution) of upper air meteorological data, especially with respect to temperature soundings, which were made on an hourly basis by aircraft. In the LAPPES program four to six helicopter temperature profiles plus two radiosondes were taken each day. However, it was evident from the discussion of the Navajo results that spatial resolution in temperature soundings may be more important than temporal resolutions in complex terrain.

In a series of experimental studies at the TVA Kingston and Johnsonville Steam Plants plume SO_2 concentration data taken from flights of fixed-wing aircraft were used by Nilsson (1975) in a preliminary analysis, to calculate σ values, employing three different methods. The first method is a statistical approach in which the Gaussian curve is approximated by a series of straight lines by taking several points on the graph. The second method is the area under a curve approach, where σ (area) is obtained by integrating the Gaussian curve equation. The third method is a peak-based approach whereby the curve peak is measured and the standard deviation is obtained by dividing the calculated width between the points where the ordinates of the graph are a certain fraction of the peak. Linear regression is performed on the σ_y values calculated by one method versus the σ_y values produced by another. For the Kingston Power Plant σ_y (stat) is greater than σ_y (area) in all cases, but when the SO_2 data taken near the Johnsonville Plant are adjusted to account for instrument time lag (this data adjustment procedure is not done for the Kingston graphs), the resulting regression showed a slope much closer to unity. These results are summarized in Table 18. The study did not focus on plume-terrain effects at all in the program, but was primarily concerned with the application of different statistical tests on plume location and dimensions. An analysis of the Kingston data by TVA is in progress.

TABLE 18. σ_Y VALUES AS OBTAINED BY NILSSON (1975) AS WELL AS CORRELATION COEFFICIENTS AND SLOPES OF THE REGRESSION CURVES

	Kingston	Johnsonville		
	Area vs Stat	Area vs Stat	10 Percent vs Stat	10 Percent vs Area
σ_Y	396,612	489,420	490,415	415,414
Correlation Coefficient	.716	.879	.963	.915
Slope	.464	.755	.816	.914

In the GEOMET analysis of Kingston data, terrain characteristics were determined using elevations read from USGS topographic maps for 200 m increments between the source and the six SO_2 monitoring stations. Figure 19 shows elevation cross-sections from the Kingston Steam plant to six monitoring stations, together with SDE values. Ground-level SO_2 measurement stations were labeled from 1 through 6. Data were analyzed for the following time periods.

Station No. 1	1/72 to 6/72, 1/74 to 12/74
Station No. 2	1/74 to 12/74
Station No. 3	1/74 to 12/74
Station No. 4	1/74 to 12/74
Station No. 5	1/74 to 12/74
Station No. 6	7/72 to 12/72, 1/74 to 12/74.

Wind speed, wind direction and temperature measured at about 46 m above the ground, and temperature measured at 1.22 m above the ground, were available in the Kingston data. Wind direction was coded in 16 compass directions. Stack parameters were obtained from FPC Form 67. Gas temperature and exit velocity were calculated from daily coal consumption rates, BTU content of coal, and BTU rating of the unit. SO_2 emission rates were provided by TVA in tons per day for each unit.

An analysis was made by GEOMET between measured concentrations and those estimated using a standard Gaussian plume dispersion model for point sources. The dispersion model is a multiple-source Gaussian plume model, adopted from EPA's UNAMAP (Users' Network for Applied Modeling of Air Pollution). Plume rise is calculated according to Brigg's plume rise estimates. The program was modified to use dispersion parameters σ_y and σ_z, based on the TVA curves (TVA 1970). Temperature gradient measurements were used to determine the closest stability class for the observation period; no unstable condition (less than -0.13°K/100m) was included in this analysis.

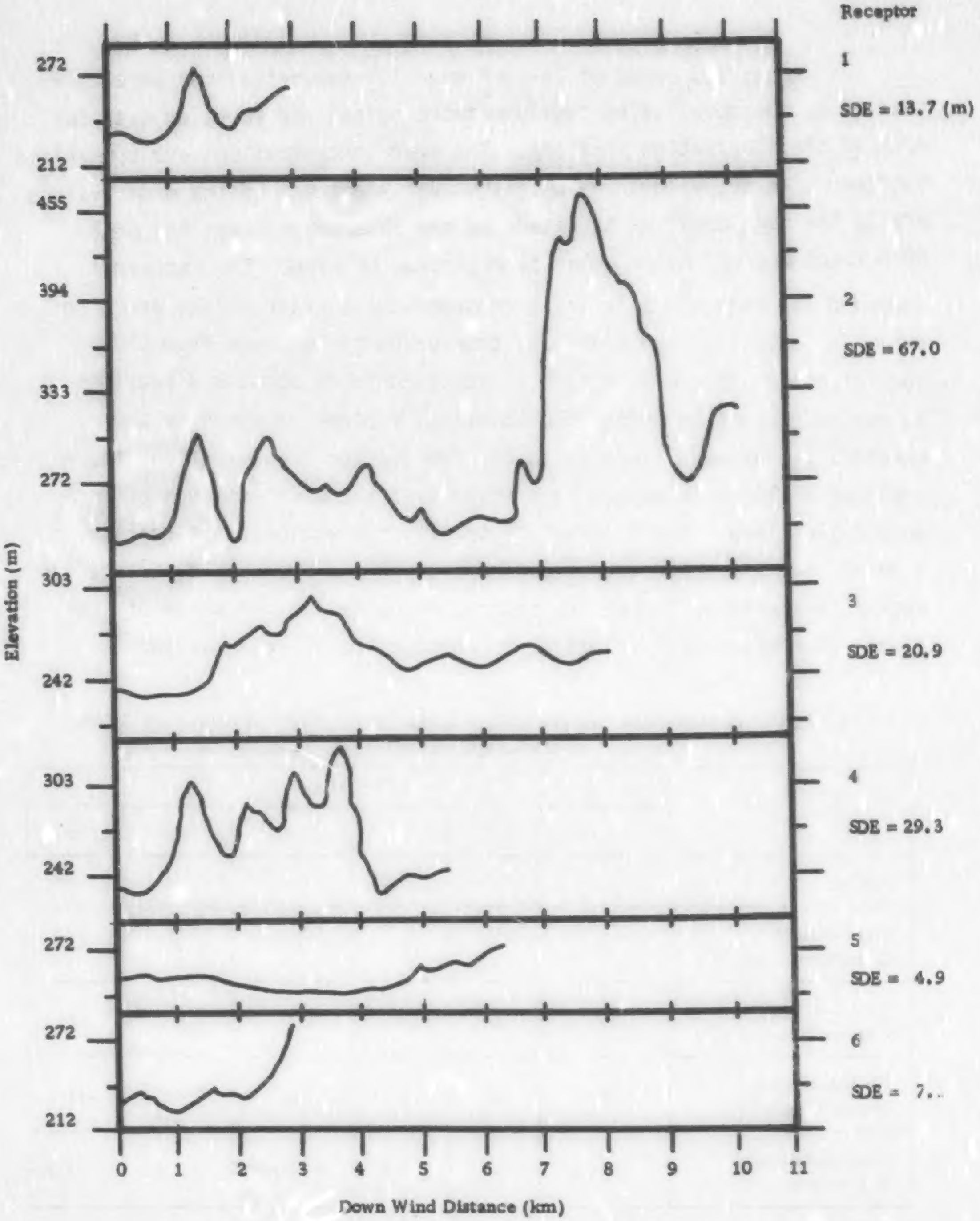

Figure 19. Elevation Profile from Kingston Plant to Each Receptor

About 125 cases of 24-hour mean SO_2 concentrations were calculated by the model using hourly meteorological and emission data for each of six observation stations. The mean concentrations are summarized together with SDE values in Table 19. Fifty percent of the model values are in the same order of magnitude as the observed values; the other 50 percent are off by an order of magnitude or more. The ratios of computed concentrations to the corresponding observed values are also shown in Table 19. The principal conclusion to be drawn from these comparisons is that wind direction resolved to 16 compass directions is not sufficient to define the location of plume relative to the monitoring stations. Between adjacent elevation cross-sections there could be hills or valleys. This emphasizes the short-comings of computing SDE along a small number of specific directions. In reality, a plume travels in the downwind direction in a narrow-angle sector; and terrain features in the entire sector influence the diffusion of the plume. Therefore, it is logical to compute the SDE of a sector.

TABLE 19. COMPARISON OF COMPUTED AND OBSERVED MEAN GROUND-LEVEL SO_2 CONCENTRATIONS*

	Station					
	1	2	3	4	5	6
SDE (m)	13.7	67.4	21.0	29.3	4.9	7.0
Mean Computed Concentration ($\mu g/m^3$)	63	0	56	1	176	22
Mean Observed Concentration ($\mu g/m^3$)	237	12	17	13	20	63
Computed Minus Observed ($\mu g/m^3$)	-174	-12	39	-12	156	-41
Computed Divided by Observed	0.27	-	3.3	0.08	8.8	0.35

* About 125 cases were used for each station.

Correlations between SDE values and the values of χ_{calc}/χ_{obs} are not evident. In addition to the definition of wind direction utilized, and the deficiency in the computational method of the SDE, the reasons for the poor correlation can be (1) the terrain induced aerodynamic effect is not included, and (2) the small amount of data in the preliminary GEOMET analyses suggest that a plume dispersion model for complex terrain requires at least the following capabilities:

1. Inclusion of a means of categorizing terrain numerically by sectors and by distance from the source.
2. Inclusion of wind flows of varying pattern rather than just a uniform flow.

SUMMARY OF FIELD STUDIES

Table 20 (Part A, Plume Sampling Characteristics and Part B, Meteorological Data) is a compendium of the 16 field studies that were reviewed in Section IX. The major objective of each of these research efforts has been the compilation of data regarding the physical behavior of either stack plumes or tracer material in complex terrain. The format of this summary includes (in Table 20A) a statement of study title and investigators, the nature of pollutant (tracer) and source, and a brief project comment. The information matrix categorically depicts the nature of the field observations for each study, describing the characteristics of ground and airborne sampling efforts. The details concerning ground sampling include the number of samplers, the distance range, the terrain characteristics of samplers and the number of sampling days. The distance range, number of sampling days and the sampled plume characteristics are listed for airborne sampling. Meteorological observations are portrayed in Table 20B with information concerning methodology of temperature measurements, type and range of aerial turbulence measurements, frequencies of stable, neutral, or unstable stabilities and type of wind data along with range of wind speeds and wind direction characteristics.

TABLE 20. SUMMARY OF FIELD STUDIES OF TRANSPORT AND DIFFUSION IN COMPLEX TERRAIN FROM ELEVATED SOURCES

A. Plume and Sampling Characteristics

Study Number	Study Title and Investigators	Nature of Pollutant (Tracer) and Source	Comments	Ground Sampling				Airborne Sampling		
				Number of Samplers	Distance Range	Terrain Characteristics at Samplers	Number of Days	Distance Range	Number of Days	Plume Characteristics Sampled
1	"Particulate Diffusion Over Irregular Terrain," McMullen and Perkins 1963	Fluorescent particles released from straight line airplane flight	Some of the earliest FP diffusion trials ever conducted in complex terrain	≈ 100	11-21 km	450-600 m above valley floor	4	27 km	4	Downwind and return to release line
2	"Diffusion Over Coastal Mountains of Southern California," Hinds 1970	Releases from ZnS from ground-level point sources	Diffusion comparisons over mountains and flat terrain show nighttime stable conditions lead to interactions between terrain-induced drainage flow and synoptic scale flow in complex terrain	43	≈ 8 km	30-500 m above sea level	113 tests	-	-	-
3	"Plume Dispersion Studies and Modeling for Fossil-Fueled Power Plants," Weil 1974	SO_2 plume from Dickerson Power Plant	Study clearly demonstrated increased turbulence due to rough terrain	Mobile van (1)	Unknown	Unknown	Oct 1972 thru Apr 1973 (no. of days not known precisely)	-	-	-

TABLE 20. (CONTINUED)

A. Plume and Sampling Characteristics										
				Ground Sampling				Airborne Sampling		
Study Number	Study Title and Investigators	Nature of Pollutant (Tracer) and Source	Comments	Number of Samplers	Distance Range	Terrain Characteristics of Samplers	Number of Days	Distance Range	Number of Days	Plume Characteristics Sampled
4	"An Initial Evaluation of Sulfur Dioxide Measurements Conducted at Naughton Power Plant," Spangler et al. 1973	SO_2 from power plant stack	During stable flow the plume remained aloft with no ground-level impaction upon the elevated terrain downwind of the plant	3	1-5 km	≈100-250 km above stack base	6	0-15 km for extremely stable day, not known for other five days	6	Crosswind
5	"Airborne SO_2 Measurements at Four Corners Power Plant," Niemann 1973	SO_2 from power plant stack, comparison with NOAA model results	Turbulence dissipation measurements indicated enhanced dilution over complex terrain than that expected over flat land	None	-	-	-	Up to 45 km	4	Ascending spiral, crosswind, along wind
6	"Analysis of Plume Data - Four Corners Power Plant," Smith and Anderson 1974							Up to 18 km	4	Crosswind and vertical
7	"Nighttime Tracer Studies of Power Plant Plumes at the Navajo Generating Station," Hovind et al. 1973	Visible (oil fog) smoke and fluorescent particle tracers	Two cases where FP releases under stable nighttime conditions did not support the theory of direct center-line impingement on elevated terrain	85	Up to 45 km	≈ up to 1000 m above stack base	4	-	-	-
8	"Daytime Plume Simulation Tracer and Climatology Program at the Navajo Generating Station," North American Weather Consultants 1974	Visible (oil fog) smoke released upwind of elevated terrain. Fluorescent particle tracer to simulate SO_2 released by aircraft at stack and effective stack heights.	The greatest sampling network impacts were noted under light wind looping conditions on summer days	75	Up to 10 km from plant or release point	≈ up to 600 m above stack base	7	-	-	-

(Continued)

TABLE 20. (CONTINUED)

A. Plume and Sampling Characteristics

Study Number	Study Title and Investigators	Nature of Pollutant (Tracer) and Source	Comments	Ground Sampling				Airborne Sampling		
				Number of Samples	Distance Range	Terrain Characteristics of Samplers	Number of Days	Distance Range	Number of Days	Plume Characteristics Sampled
9	"Navajo Generating Station SO_2 Field Monitoring Program," Rockwell Int., MRI, SAI 1975	SO_2 from power plant stack	(Same as Study No. 8)	26	Up to 30 km	≈ up to 600 m above stack base	145 days total, 80 non-zero observation days	Up to 28 km	144	Crosswind, along wind
10	"Summary Report on Meteorological Power Plant Plume Simulation Tracer Studies in Huntington Canyon, Utah," Hovind et al. 1973a	FP and smoke releases from estimated effective stack height, by skywriter, in addition to helicopter smoke releases in area inaccessible by plane *	Extensive supplemental observational information including photographs of various types of plume behavior - much of subjective information reported valuable to understanding flow in complex terrain - several physical mechanisms for the increase of lateral dispersion observed	86 rotorod stations	Up to 15 km from plant site*	Up to 850 m above stack base*	7	Sampling was made, but details not discussed	-	-
11	"The Influence of Rough Mountainous Terrain Upon Plume Dispersion from An Elevated Source," Hovind et al. 1974	Fluorescent particles at one site and oil fog dispersion at two sites		Used at FP site only Number not provided	Up to 25 km	Unknown	6	Up to 25 km	13	Crosswind and centerline
12	"Diffusion in Canyon with Rough Mountainous Terrain," Start et al. 1975	SF_6 tracer from stack, elevated terrain, and canyon floor. Also oil fog flow.		40	0-10 km	Up to ≈ 350 m above canyon floor	11	10 km	9	Centerline
13	"Effluent Dilutions Over Mountainous Terrain," Start et al. 1974	SF_6 tracer released from smelter stack		≈ 40	0-5 km	Up to ≈ 850 m above smelter level	7	Up to 6 km	7	Centerline

(Continued)

* Refers to Tracer Program III, the most extensive of seven separate field studies (each of about one week's duration), conducted by NAWC at the Huntington Canyon Plant Site.

TABLE 20. (CONTINUED)

A. Plume and Sampling Characteristics

Study Number	Study Title and Investigators	Nature of Pollutant (Tracer) and Source	Comments	Ground Sampling				Airborne Sampling		
				Number of Samplers	Distance Range	Terrain Characteristics of Samplers	Number of Days	Distance Range	Number of Days	Plume Characteristics Sampled
14	"Elevated FP Tracer Releases from Kaiparowits Power Plant Site," North American Weather Consultants 1974	FP tracer released from aircraft at two sites supplemented by oil fog releases	Vicinity of proposed plant site subject of tracer studies and model simulation-comparisons	Not stated precisely, > 20	-	-	Not stated but total of 10 tracer releases made	Unknown	-	-
15	"Large Power Plant Effluent Study - Vol. 4," Schiermeier 1971	SO_2 from power plant stack	Airborne sampling data provide large data base for future studies of terrain influences	Up to 14	Variable - up to 27 km	Up to mid-stack height	27	Up to 16 km	175	Crosswind at several altitudes
16	TVA Field Smelters in the Vicinity of Kingston Power Plant, 1973-1975	SO_2 from power plant stack		11 fixed sites, plus portable bubblers	Up to 10 km	Up to 250 m above the plant grade	Nov-Dec 1973, Mar-Apr 1974, Jan-Mar 1975	Up to 21 km	Unknown	Crosswind at several altitudes

TABLE 20. (CONTINUED)

Study Number	B. Meteorological Data									
		Turbulence		Observed Stability Conditions				Wind		
	Temperature	Types of Measurements	Range of Aerial Measurements	Frequency of Stable	Frequency of Neutral	Frequency of Unstable	Other Stability Information	Types of Wind Data	Range of Wind Speeds	Wind Direction Characteristics
1	Aircraft Profiles Sampling at ground and tower	-	-	-	-	-	-	Radiosonde, tower, pibal, surface	Light (3) Strong (1)	Predominantly SSE
2	Hourly radiosondes Sampling at surface and 100 m	-	-	-	-	-	Not known, precisely, but mostly neutral and unstable	Radiosonde, tower, surface	Unknown	Unknown
3	Aircraft Profiles 2 to 3 Radiosondes Per Day	Lateral (σ_θ)	-	-	-	-	Not precisely known, but majority of sampling was probably conducted under neutral or unstable conditions (between 10am and 4pm only)	Radiosonde, tower, pibal	-	-
4	Aircraft Profiles	-	-	-	-	-	Not exactly known, but stated that plume tracking over 6-day period covered a variety of meteorological conditions including 1 day with extremely stable air mass.	Pibal, surface	Not known	Not known, but both easterly and westerly flow sampled

TABLE 20. (CONTINUED)

Study Number	B. Meteorological Data									
		Turbulence		Observed Stability Conditions				Wind		
	Temperature	Types of Measurements	Range of Aerial Measurements	Frequency of Stable	Frequency of Neutral	Frequency of Unstable	Other Stability Information	Types of Wind Data	Range of Wind Speeds	Wind Direction Characteristics
5	Aircraft Profiles	Dissipation Rate	Yes, sfc. to plume top	2	1-1/2	-	1/2 (Transitory inversion breakup)	Radiosonde, surface	Nov. cases moderate Jan. cases calm to very light	Steady easterly variable
6	Aircraft Profiles	Dissipation Rate	Yes, sfc. to plume top	4	-	-	-	Pibal	2 days light, 2 days moderate	Generally easterly near surface
7	Aircraft Profiles Ground Sampling	-	-	4	-	-	-	Pibal, stack top, surface	Very light	Great variability
8	Aircraft Profiles	Vertical	Not precisely known	-	-	-	7	Pibal, stack top	Moderate to strong	Mostly southwest
9	Aircraft Profiles 1 Radiosonde Per Day	-	-	-	-	-	Not known precisely, but majority of cases stable or neutral	Radiosonde, pibal, stack top, surface	Wide range	Observations divided into 3 categories - 2 distinct directions and remainder
10	Aircraft Profiles Ground Sampling	Dissipation Rate	-	7	-	-	-	Pibal, stack top, surface	0 to 4 m/sec	Mostly east-southeast, westerly on one occasion
11	Aircraft Profiles	Dissipation Rate	Yes to top of plume	15	4	0	-	-	Light to moderate	Unknown
12	Sampling at 3 levels on 183 m stack	-	-	5	5	1	-	Tower, stack top, surface	Light	Mostly southeast and north at stack top
13	Ground Sampling 1 Radiosonde Per Day	-	-	-	-	7	-	Radiosonde, pibal, surface	Light to moderate	All between northwest and east-northeast
14	Some vertical temperature profiles taken, but number and method unknown	Unknown	Unknown	-	-	-	Not precisely known, but tests during neutral, stable, and slightly unstable conditions	Pibal	Unknown	Unknown

(Continued)

TABLE 20. (CONCLUDED)

Study Number	B. Meteorological Data									
		Turbulence		Observed Stability Conditions				Wind		
	Temperature	Types of Measurements	Range of Aerial Measurements	Frequency of Stable	Frequency of Neutral	Frequency of Unstable	Other Stability Information	Types of Wind Data	Range of Wind Speeds	Wind Direction Characteristics
15	Aircraft Profiles Ground Sampling 2 Radiosondes Per Day	In plume turbulence measurements by MRI as part of LAPPES in spring and fall of 1968	-	Measurements made during stable morning and inversion breakup for each date				Radiosonde, pibal, surface	Light to moderate	Southwest and west dominant
16	Aircraft Profiles Sampling at 1, 2, and 48 m levels	Dissipation Rate	Yes, up to 5000 m	-	-	-	Unknown	Tower, pibal, surface	Unknown	Unknown

Section X

RECAPITULATION AND COMMENTARY

The analysis of the scope of previous studies of plume (or simulated plume) behavior in complex terrain, and its relationship to meteorological, emission, and topographical factors has revealed that a modest data base, of both theoretical and observational information has been gathered relative to important phenomena associated with flow in complex terrain. Financial restraints in conjunction with other practical considerations have dictated that field measurement programs each focus on a small number of specific objectives. The diversity of geographic sites selected, the variety in the experimental methods utilized, and the differences in the analysis of results present a formidable undertaking if the goal is to consolidate the outcomes of each of these programs into a flexible, yet coherent, universal set of rules for plume diffusion in complex terrain. However, certain common denominators have been found to link the diversity of research projects in the literature, thus allowing a number of qualitative generalizations regarding plume diffusion and transport in complex terrain to be presented.

ENHANCED TURBULENCE

Plume studies have indicated that an expected increase in turbulence exists in complex terrain over that found over relatively flat surfaces. This conclusion has been achieved through analyses of wind direction fluctuations, comparison of σ_y and σ_z measured values to the Pasquill-Gifford values applicable to flat terrain, and by model-to-measurement comparisons of effluent or tracer concentrations. As earlier references have made clear, a major purpose of many field investigations has been the collection of plume geometry data from which inferences can be made concerning the levels of turbulence present near power plants located in complex terrain. It appears from a study of this literature that in general, aircraft measurements of plume cross-sections yield the best information about plume geometry. When aircraft measurements are not available, comparisons of measured ground-level concentrations to model calculations of ground-level concentrations in flat terrain may be used to estimate turbulence enhancement due to the complex terrain.

TERRAIN IMPINGEMENT

One of the important questions to be approached is how stability affects the proximity of the plume to the surrounding complex terrain. For those studies in which the stability of the atmosphere was reported in detail, the stable conditons most frequently characterized the cases studied, principally because study objectives often were to examine the validity of hypothesized NOAA model plume impingement on elevated terrain under these conditions. The Navajo studies (Rockwell, MRI and SRI, 1975) contained two cases where the few FP releases under stable nighttime conditions did not support the theory of direct centerline impingement on elevated terrain. Even though the tracer was initially transported towards critical elevations (between stack top and effective stack height), no distinct maximum impact zones were indicated, except for one case, and that was for a location below the release elevation (however, the highest SO_2 ground concentrations occurred on the cliffs). Evidently, strong vertical mixing was occurring near the elevated terrain even with the stable conditions, probably as a result of strong wind shear induced by the rising terrain. Direct impingement upon elevated terrain was also not evident in the Huntington Canyon studies for stable conditions;* when the plume was directed toward the canyon walls, the observations indicate that it was deflected away from the wall before impingement.* During lapse conditions at Huntington Canyon it was concluded that transient impactions on the order of several minutes may indeed occur at any point, however the results are limited in significance; one reason being because the topography of the release points varied greatly for the differing meterorological conditions. Direct impingement of a plume against steep canyon walls was observed by Start et al.

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* The lone stable release from an elevated position on a natural prominence visually showed the plume to impinge on the canyon wall, however, it moved up the slope, away from the samplers, so no quantitative data were obtained. Kinematically produced deformation effects may have been responsible.

in one Stability B test case near the Garfield Smelter. In the Navajo studies, oil smoke releases during transitional fumigation conditions allowed transport of the smoke into the rills of one of the elevated terrain features (Leche-e Rock) 8 km downwind; earlier stable conditions had presented no evidence of direct centerline impingement. The turbulent downward transport of the plume was expected to result in the greatest impact on elevated terrain during transitional fumigation periods between stable morning and neutral unstable afternoon conditions. The greatest Navajo sampling network impacts were noted under light wind looping conditions on summer days. Most of the field studies have been of limited duration (generally less than two weeks, and in most cases of the order of one week), and as a result have examined dispersion over a relatively limited range of atmospheric stability conditions. However, plume behavior in complex terrain during documented transitory conditions (especially inversion breakup) has been investigated and discussed in detail (i.e., over periods greater than one week) in the six-month Navajo study by Rockwell International, Meteorology Research, Inc., and Systems Applications, Inc. (1975). Many of the observed impingement events occurred during unstable conditions and primarily at the initiation of unstable conditions. Temperature soundings atop a relatively flat, elevated terrain feature for the days on which the plume passed close to or intersected that terrain (resulting in the highest observed ground concentrations) were not conducted, but soundings near the plant suggested that fumigation rather than stable conditions may have resulted in the high concentrations observed.

PLUME DIMENSIONS

There is some evidence from the LAPPES program (Schiermeier 1970, 1972), GEOMET's analyses of the LAPPES data (1976), and the Four Corners studies (Niemann 1973, Smith and Anderson 1974) that horizontal spreading (i.e., σ_y) is amplified to a greater extent than vertical spreading in complex terrain, particularly under stable conditions. The resultant increase of σ_y has been attributed primarily to plume separation processes and plume meandering which occur when the higher terrain elevations are confronted by the plume. Plume meandering was frequent in the Navajo region under stable light conditions due to gradient flow interaction with the nocturnal thermal circulations induced by terrain. Both Smith and Anderson and Niemann concluded that significant differences in plume dimension and dilution could occur without any major change in the stability category as determined by turbulence variations at the plume level. This indicates that perhaps a more concise way of characterizing dispersion other than atmospheric stability may be needed in complex terrain. At Huntington Canyon, moderate to heavy turbulence conditions have been documented with stable conditions, with enhanced dilution as stabilities increased. Recent acoustic sounding data shows extensive waves and breaking waves in stable layers, thus indicating that stable layers are not quiescent. The cooling and subsequent development of radiation inversions in irregular topography cause drainage of air down the slopes, and this acceleration of air has to set off compensating circulations. Thus, inversions and their possible subsequent anti-mountain circulations can be said to enhance turbulence in this complex terrain situation. Studies have also shown turbulence levels in the down-valley and downslope flow to increase as a function of the inversion intensity, probably due to the role of inversions in the production of katabatic winds. Lee wave phenomena have also been pointed out as responsible for increased turbulent effects, and mechanical turbulence and vertical shear are postulated to arise over and around peaks and ridges of mountainous terrain. As was evident in the Navajo program, significant interactions between terrain-induced drainage flow and the prevailing gradient flow aloft may occur near elevated features.

PLUME CENTERLINE CONCENTRATIONS

A comparison study of measured aerial plume centerline SO_2 concentrations to the concentrations predicted by the Gaussian plume model on one test day characterized by stable conditions near the Naughton Plant site showed results similar to those obtained near the Four Corners and Navajo plants for stable conditions. Within 2 km of Naughton for the one case situation described by Spangler et al. (1973) model SO_2 centerline concentrations were greater than seven times those observed; while beyond 7 km the calculations were within a factor of 2 of the measured values. At Four Corners (Smith and Anderson 1974) 11 data points representing three test days were overpredicted by factors of 5 to 30 at 1 km from the stack; the measured/calculated values were within a factor of 2 beyond 12 km, and results were similar at 18 km from the plant. Niemann's (1973) comparison of Four Corners measured instantaneous SO_2 plume centerline concentrations for six flights under neutral and very stable conditions in two different seasons support the Spangler et al. (1973) and the Smith and Anderson (1974) results. For the neutral condition data, the conventional plume model for centerline dispersion was used for comparison, and for the stable data, the NOAA model predictions were used. Both models overestimated the measured concentrations by a factor of 10 under both stability conditions close to the plant. At distances greater than 15 km, the conventional Gaussian calculations generally agreed with the observed values, while the NOAA model over-predicted centerline concentrations by a factor of 2 under stable conditions. Seven comparisons of equivalent maximum hourly ground concentrations obtained from airborne and stacktop FP releases and the calculations of the NOAA centerline prediction equation were made at Navajo (Hovind et al. 1973 draft report). Under stated stable conditions, model overestimations ranged from 20 to 300 times the actual observed SO_2 values. At Huntington Canyon, 11 test comparisons between observed plume centerline concentrations and corresponding values calculated by the standard Gaussian plume model showed minimal differences during moderate to strong temperature lapse; canyon dilutions of 5 times to 15 times that of "standard" curves were evident for strong inversions (Start et al. 1975). In the Hovind et al. (1973c) study in the same region, the NOAA model overestimated centerline concentrations by 10 times at 2.5 km downwind, but approached actual measured values beyond 10 km during one week of observations in which all except one of the days were characterized by stable conditions; the remaining day was neutral. The Hovind et al. (1974) study, compared the ratio of calculated standard Gaussian plume centerline concentrations to observed values at three sites with different roughness characteristics. These ratios ranged from 10.2 for the Huntington Canyon site under F stability for six cases, to 6.0 for four cases of D stability in the rugged open canyon site, to 1.2 for nine cases of E stability in flat open terrain. Near the Garfield Smelter, Start et al. (1974) found that for this location, characterized by complex air circulations due to the presence of the nearby Great Salt Lake, measured $\chi u/Q$ values were 2 to 4 times less than predicted by standard curves in seven elevated FP releases in lapse conditions. INTERA (1975) verifies these results in three tests which found NOAA model concentrations to be greater than observed centerline values by factors of 2 to 5 (for stability categories of B and C) at distances within 4 km of the plant in two of the three tests, and for all distances in the last test. Beyond 4 km, calculated values approached observed values. Hoffnagle et al. (1975) compared the measured data of four FP tests at two proposed plant sites in complex terrain to NOAA model calculations where one test was characterized by stable conditions, and the other three characterized by unstable conditions. The mean ratio of calculated to observed values in all four cases (7 to 20 receptors evaluated in each case) was within 30 percent of unity.

Comparisons of the NOAA, EPA, and INTERA models systematically show greater overestimation with respect to plume centerline values close to the Navajo, Garfield, and Kaiparowits sites than at greater distances, suggesting that neither the Gaussian nor INTERA models in these cases adequately take into account the initial dilution due to emission-related parameters (buoyancy and turbulence generated by stack exit velocity and possible immediate terrain effects). The validity of the comparisons for aerial data may be limited because of the different averaging times representing the model calculations (one hour, 24 hours for Valley) and measurements (one minute). The effective plume height used in the model calculations was determined from aerial data, rather than from plume rise formula; thus, the true predictive capability of these differing formulae could not be realistically assessed for the given meteorological and topographical conditions.

STATE-OF-THE-ART FOR MODELING TRANSPORT AND DIFFUSION OF CONTINUOUS ELEVATED PLUMES IN COMPLEX TERRAIN

Comparisons of measured plume dimensions to the Pasquill-Gifford diffusion parameters (σ_y and σ_z) in complex terrain indicate that, for both E and F stabilities, rapid growth of the plume occurs initially followed by slower growth than predicted by the standard Gaussian plume model. The model values in general underestimate initial lateral spread, and overestimate vertical spread at greater distances.

Several studies have revealed that greater discrepancies between modeled/observed concentrations occur under stable conditions than under neutral conditions. The NOAA Huntington Canyon study, the Niemann 1973 study at Four Corners, and the NOAA model/measurement comparisons made by INTERA (1975), for Navajo (under stable conditions), Kaiparowits (unstable conditions) and Garfield (unstable conditions) support this conclusion.

Analysis of the available numerical models developed or adapted for use in complex terrain from the literature reveal no evidence of validation for the Environmental Research and Technology (Egan 1975) modified Gaussian model or the Aerovironment (MacCready et al. 1974) modified Gaussian model. Research has been done in the past on effects of topography on airflow (with two outstanding examples being drainage-counter-drainage winds and hydraulic "jump" flow) but nothing surfaced during this review that revealed an attempt to develop and validate a point source air quality model to incorporate these effects. Current models are incapable of simulating plume behavior when the lower part of the plume is caught in drainage flow and the upper part of the plume is in the counter flow. There exists a need to determine how severely downwash conditions affect the performance of the available models. Presently available models represent the influences of terrain by a receptor height adjustment procedure, or in the case of the INTERA model, by means of a wind speed profile. The detailed turbulence influences of terrain characteristics and terrain obstructions are required for reliable estimations of air quality impact; future emphasis must be placed on relating terrain roughness to dispersion in complex terrain in a quantitative manner.

Appendix A

BIBLIOGRAPHY ON NONCONSERVATIVE PLUME BEHAVIOP

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Appendix B

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16. ABSTRACT

This report reviews the literature of scientific studies of the behavior of stack plumes from fossil-fueled electric power plants in complex (hilly or mountainous) terrain. Non-conservative chemical transformation and depletion, and conservative transport and diffusion of pollutants are considered. Studies of SO_2 oxidation rates in power plant plumes are described and the primary mechanisms for conversion to sulfate are detailed. Scavenging of SO_2 from plumes by precipitation is reviewed along with surface contact and deposition as important plume depletion processes. Current theories of airflow, turbulence and diffusion phenomena in complex terrain are described, and are exemplified through the review of sixteen field observation programs of the physical behavior of plumes from continuous elevated sources in complex terrain. The review discusses program objectives, data sampling methodologies, model-to-measurement comparisons associated with the field program conclusions. The types of models available to simulate plume behavior numerically are discussed in the contexts of these programs. In addition, results are reported of an independent GEOMET analysis of σ_y and σ_z relationships in the Gaussian plume model over complex terrain, using data from LAPPES and TVA field measurement programs.

delta(Y) delta(Z)

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